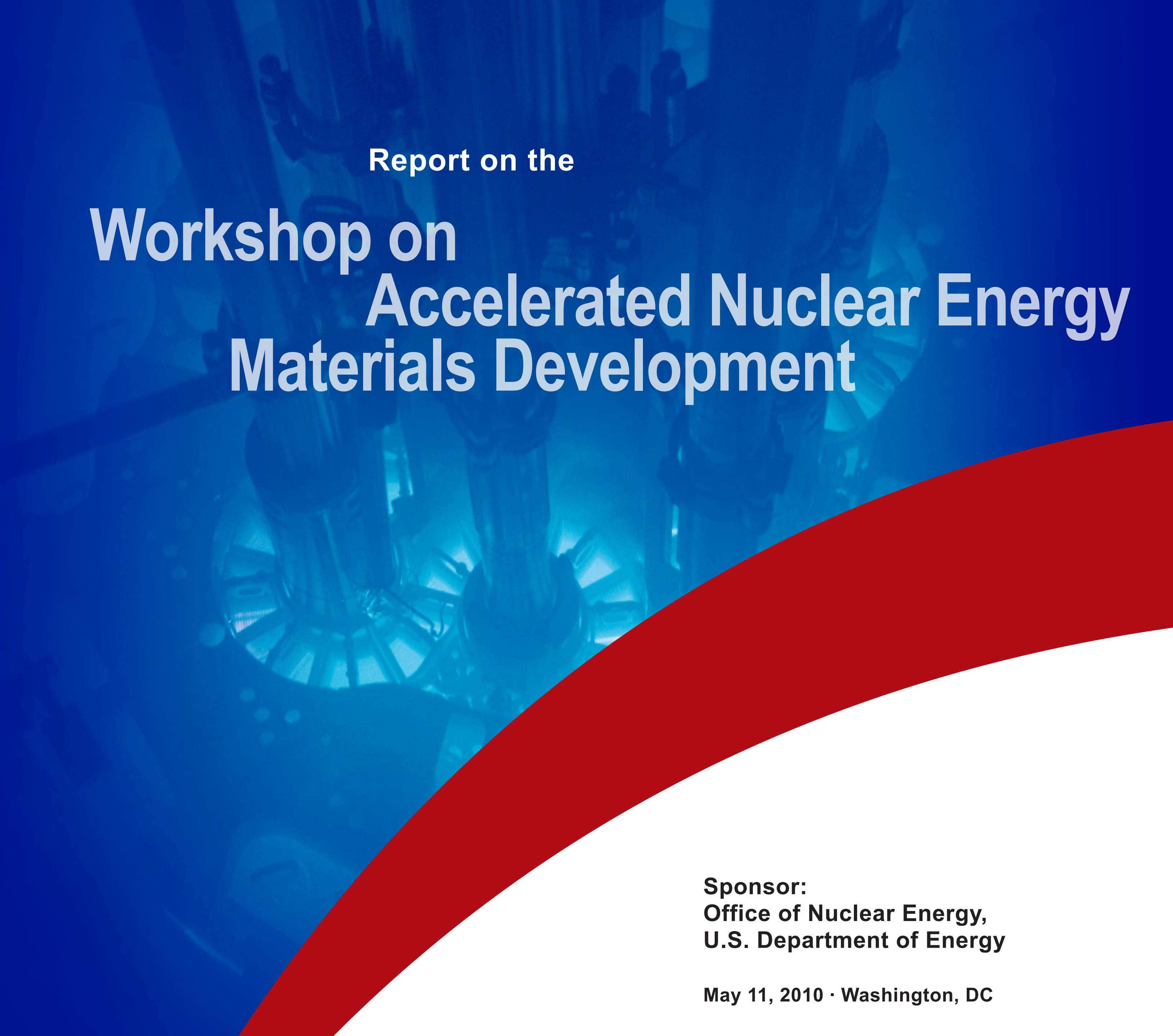

Report on the

Workshop on Accelerated Nuclear Energy Materials Development

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About the Cover

The Advanced Test Reactor at Idaho National Laboratory has a unique serpentine fuel arrangement, which allows researchers to simultaneously test materials under varied conditions. The characteristic blue glow is due to Cerenkov radiation, which is emitted when a charged particle such as an electron passes through the cooling water at a speed faster than the speed of light in the water. (Reprinted courtesy of Idaho National Laboratory, http://www.flickr.com/photos/inl/3852591286/.)

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Contents

Executive Summary 1
1. Introduction 3
2. Uncertainty Quantification 23
3. Theory, Simulation, and Modeling 29
4. Ion-Beam Capabilities and Limitations 32
5. Topical Area 1: Model Development 35
6. Topical Area 2: Chemistry and Property Evolution of Fuels 37
7. Topical Area 3: Advanced Clad Materials Development 39
8. Topical Area 4: Fuel-Clad Interaction Leading to Fuel Failure 42
9. Topical Area 5: Design a Radiation-Tolerant Material 44
10. Stakeholder Discussion 46
11. Interactions, Interdependencies, and Synergies with Other Organizations 56
12. Workshop Recommendations 57
13. Conclusions 59
Acknowledgements 61
Appendix A: Workshop Agenda 62
Appendix B: Questions and Answers 63
Appendix C: Workshop Participants 95
Appendix D: Facilities and Other Resources 96

Executive Summary

This document reports on the Office of Nuclear Energy's (NE's) Workshop on Accelerated Nuclear Energy Materials Development held May 11, 2010, in Washington, DC. The purpose of the workshop was twofold: (1) to provide feedback on an initiative to use uncertainty quantification (UQ) to integrate theory, simulation, and modeling with accelerated experimentation to predict the behavior of materials and fuels in an irradiation environment and thereby accelerate the lengthy materials design and qualification process; and (2) to provide feedback on and refinement to five topical areas to develop predictive models for fuels and cladding and new radiation-tolerant materials. The goal of the workshop was to gather technical feedback with respect to the Office of Nuclear Energy's research and development while also identifying and highlighting crosscutting capability and applicability of the initiative to other federal offices, including the Department of Energy's (DOE's) National Nuclear Security Administration (NNSA), Nuclear Regulatory Commission (NRC), DOE Office of Basic Energy Sciences (BES), DOE Office of Fusion Energy Sciences (FES), and Naval Reactors.

The goals of the initiative are twofold: (1) develop time- and length-scale transcending models that predict material properties using UQ to effectively integrate theory, simulation, and modeling with accelerated experiments; and (2) design and develop new radiation-tolerant materials using the knowledge gained and methodologies created to shorten the development and qualification time and reduce cost.

The initiative is crosscutting and has synergy with industry and other federal offices including Naval Reactors, NRC, FES, BES, and the Office of Advanced Scientific Computing Research (ASCR). It is distinguished by its use of uncertainty quantification to effectively integrate theory, simulation, and modeling with high-dose experimental capabilities. The initiative aims to bring the methodology that is being successfully applied in NNSA's Stockpile Stewardship Program to accelerate the development of advanced nuclear energy materials. Industry, Naval Reactors, and NRC expressed support for this type of initiative and a general interest in collaboration in the area of light water reactor materials research.

A successful initiative will provide a methodology that will enable the evaluation of the performance of fuels and materials in time- and cost-effective "out of core" experiments. It will also result in a more effective use of existing reactor resources and enable the timely design and development of required new materials for advanced reactors.

Fifty-nine participants attended the workshop: 38 were from national laboratories, 4 were from universities, 8 were from industry, and 9 were from federal offices. Of those 9, 5 were from NE, 1 was from NRC, 1 was from FES, and 2 were from Naval Reactors.

Based on the questions that were asked at the workshop, the stakeholder discussion, and subsequent interactions with participants, the following conclusions have been drawn:

- The initiative should build a scientifically defendable argument for the applicability of models developed using accelerated experiments to neutron irradiation environments.
- The initiative must strive to be a nationwide network of experts and facilities at universities, industries, and national laboratories led by a lead institution and working as a team.
- This initiative should focus on accelerating materials development through the synergistic use of mechanistic high-rate experiments coupled with advanced theory, simulation, and modeling and integrated using UQ leading to the design of new fission reactor materials for the extreme of high irradiation dose and high burn-up.
- Where neutron-irradiation experiments are not feasible, either because the appropriate spectrum is unavailable or because long irradiation times would be required, ion-beam experiments should be coupled with theory, simulation, and modeling and integrated using UQ to achieve the needed advances in irradiation effects scaling.
- Models developed for materials under neutron-irradiation conditions at this extreme cannot be validated because little or no neutron-irradiation data exists. Consequently, uncertainty quantification will be particularly important in quantifying uncertainties into dose regimes that have not been explored using neutrons.

The scope of this initiative is responsive to a request from NE to focus on fuels and cladding. Five topical areas (TA) for research are discussed in this report, each of which exercises different aspects of the methodology. Related work may already be ongoing in DOE by other organizations. The topical areas are designed to demonstrate the added contribution brought by this initiative, are graded in difficulty and cost, and can be sequenced to demonstrate incremental levels of success

TA1 provides an opportunity to establish and exercise the methodology to develop a strength model that can be validated against existing neutron-irradiated material. TA2 applies the methodology in the area of thermodynamics and phase transformations of complex actinides to better understand the chemistry and property evolution of reactor fuels. TA3 extends the methodology from TA1 and TA2 but adds the complexity of irradiation creep. TA4 applies the methodology in the area of failure, moving from continuously changing properties to prediction of a failure event. TA5 applies the methodology in the area of materials design and development.

1. Introduction

Purpose

This document reports on the Workshop on Accelerated Nuclear Energy Materials Development held May 11, 2010, in Washington, DC. The purpose of the workshop was twofold: (1) to provide feedback on an initiative to use uncertainty quantification to integrate theory, simulation, and modeling with accelerated experimentation to predict the behavior of materials and fuels in an irradiation environment and thereby accelerate the lengthy materials design and qualification process and (2) to provide feedback and refinement to five topical areas to develop predictive models for fuels and cladding and new radiation-tolerant materials. The goal of the workshop was to gather technical feedback with respect to NE's research and development while also identifying and highlighting crosscutting capability and applicability of the initiative to other federal offices, including NNSA, NRC, BES, FES, and Naval Reactors.

Background

The greatest technical bottlenecks to shortening the time and reducing the cost of developing and licensing new technology for nuclear energy applications is the ability to confidently predict the behavior of materials in the extreme environments of the nuclear reactor and to develop new materials that better withstand this environment. These bottlenecks are crosscutting issues not only for thermal, light water reactors (LWRs) but also for high-temperature gas-cooled reactors and fast reactors. Overcoming these bottlenecks requires a predictive capability for materials properties and performance in extreme environments consisting of high-dose neutron irradiation, fission of actinide fuel, high temperature, high stress, and corrosive media.

Predictive modeling capabilities have been elusive because of the complexities of the materials and the nuclear reactor environment. Recent developments in theory, simulation, and modeling as well as in accelerated experiments, advanced characterization, and a new area known as uncertainty quantification (Adams et al. 2009) offer hope for a step change in our ability to understand and predict the extremes of high irradiation dose through better mechanistic understanding of phenomena and quantification of uncertainties.

In the short term, predictive modeling and accelerated experiments integrated through uncertainty quantification could be applied to existing nuclear reactors and fuel designs to help enable plant life extension and to license reactors for uprated conditions. In the long term, this mechanistic understanding of performance and degradation phenomena developed coupled with this methodology could reduce the time and cost required to develop and evaluate new materials and aid licensing of the materials.

Modeling Challenges for Irradiation Effects[1]

The effect of irradiation on materials is a classic example of an inherently multiscale phenomenon (Figure 1). The added complexity that radiation effects introduce in materials is the overarching concern for advanced nuclear energy systems. The pertinent processes that must be modeled span more than 10 orders of magnitude in size, from the subatomic nuclear to the structural component level, and 22 orders of magnitude in time, from the sub-picosecond level of nuclear collisions to decade-long component service lifetimes (Wirth et al. 2004). Many variables are needed to describe the mix of nano- or microstructural features that are formed when irradiation degrades the physical and mechanical properties of nuclear fuels, cladding, and structural materials. The most important features are the initial material composition and microstructure, the thermomechanical loads, and the irradiation history.

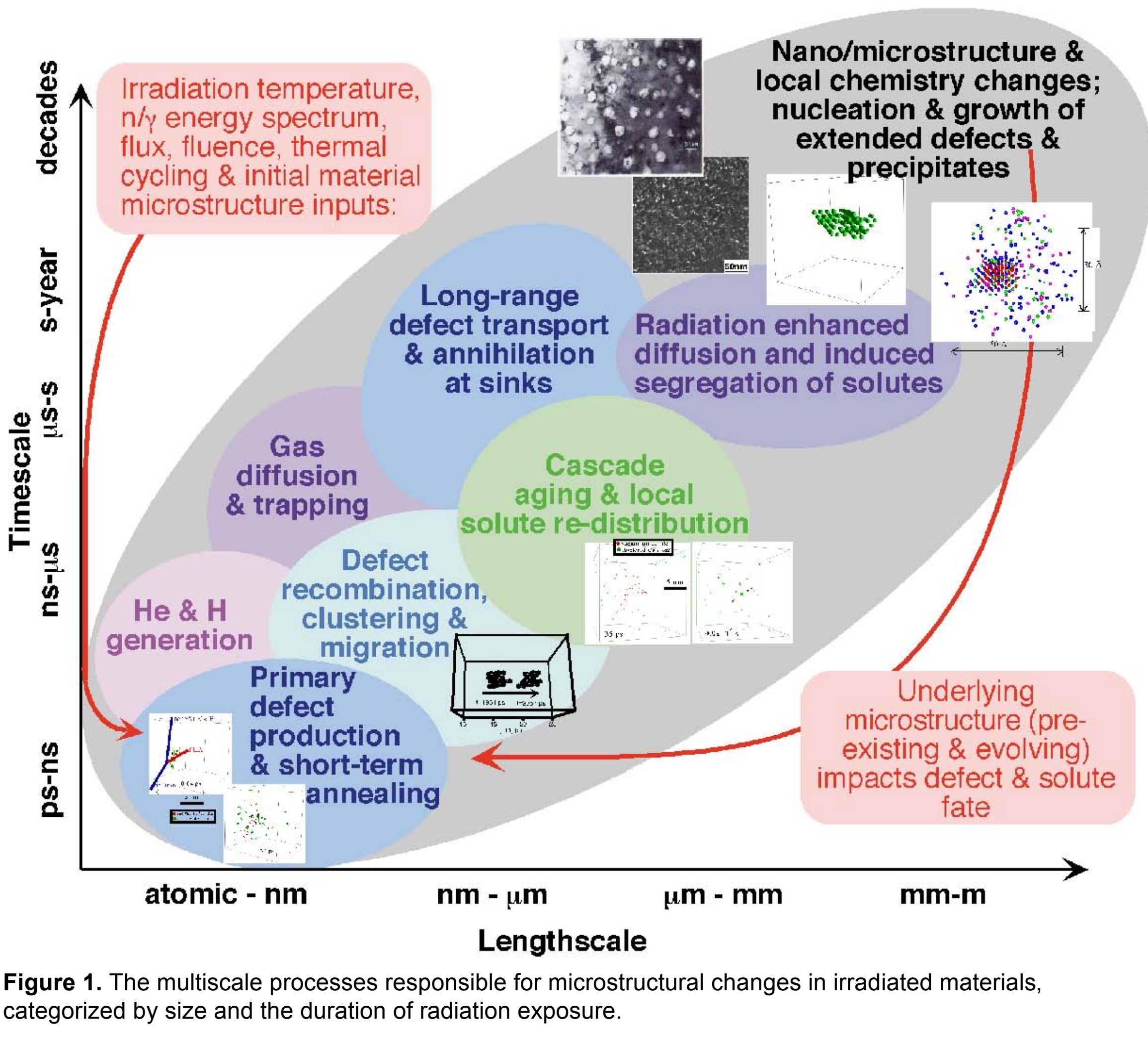

Figure 1. The multiscale processes responsible for microstructural changes in irradiated materials, categorized by size and the duration of radiation exposure.

[1] Excerpted, with permission, from Moniz, Ernest, and Robert Rosner. 2009. Scientific Grand Challenges: Science Based Nuclear Energy Systems Enabled by Advanced Modeling and Simulation at the Extreme Scale. In *ASCR Program Documents*. Crystal City, VA: Office of Science, Advanced Scientific Computer Research and the Office of Nuclear Energy.

At the smallest scale, radiation damage is continually occurring when energetic primary knock-on atoms (PKA) form through elastic collisions of high-energy neutrons with reactor materials, At the same time, radiation generates high concentrations of fission products in fuels and transmutants in cladding and structural materials that can profoundly alter the overall chemistry of materials, especially at high burn-up. The PKAs as well as recoiling fission products and transmutant nuclei quickly lose kinetic. For structural materials the electronic excitations from PKAs and their progeny are not generally thought to produce atomic defects in metallic structural materials. Subsequently, the ballistic collisions, as a result of a chain of atomic collision displacements, produce a cascade of vacancy and self-interstitial defects. For the case of fission fragments, with energies of ~100 MeV, the consequences of the plasma-like electron-hole tracks are not well understood. High-energy displacement cascades in structural materials due to PKAs occur over very short time spans of 100 picoseconds or less and in small volumes, covering a size of about 50 nm or less in length. They can be modeled using molecular dynamics (MD) simulations if accurate potentials are available.

Materials scientists who have studied the physics of primary damage production in high-energy displacement cascades using MD simulations (Calder and Bacon 1993) have found that:

- The intra-cascade recombination of vacancies and self-interstitial atoms (SIAs) results in ~30% of the defect production expected from displacement theory;
- Many-body collision effects produce a spatial correlation (separation) of the vacancy and SIA defects;
- Substantial clustering of the SIAs and, to a lesser extent, the vacancies occurs within the cascade volume; and
- High-energy displacement cascades tend to break up into lobes or sub-cascades that may also enhance recombination (Calder and Bacon 1993; Phythian et al. 1995).

Research has concluded, however, that the subsequent diffusional transport and evolution of the defects produced during displacement cascades are the primary cause of radiation effects in materials and changes in material microstructure (Wirth et al. 2004; Odette et al. 2001), in addition to solutes and transmutant impurities. Displacement cascades begin by having important spatial impacts at small scales that continue to play a significant role over much larger scales, as do processes that include defect recombination, clustering, and migration as well as gas and solute diffusion and trapping. Consequently, changes in the underlying materials structure reflect the time and temperature kinetics of diffusive and reactive processes, although they are strongly influenced by spatial correlations associated with the microstructure and the continuous production of new radiation damage.

Since there is such a wide range of timescales and a "rare-event" nature characteristic of controlling mechanisms, efforts to model the effects of radiation on materials are extremely challenging. It is often difficult to obtain even tentative characterizations of the processes. Indeed, materials scientists have been unable to create models of microstructural evolution during service that accurately consider point defects, dislocations, and grain boundaries.

Today, materials scientists face a substantial challenge: to discover the processes that control how nuclear materials perform and use them to model this behavior. To create what we would regard as high-fidelity models, scientists would need to develop a more profound understanding of irradiation effects and microstructural evolution through a combination of experimentation, theoretical analysis, and computation. Exascale computing can enable such breakthroughs through discovery-class simulations, although scientists would need to assess how accurately models can describe critical physical phenomena through uncertainty quantification. If they could overcome some of the important limitations in current knowledge about the kinetic processes that control defect cluster and microstructure evolution, as well as materials degradation and failure modes, they would open the way to include accurate descriptions of key controlling processes in high-fidelity models and reduce errors currently due to in-service surprises.

The challenges that materials scientists face in developing high-fidelity nuclear materials performance models are many and include the following:

- Bridging the inherently multiscale time and size scales that characterize materials degradation in nuclear environments;
- Dealing with the complexity of multi-component materials systems, including those in which the chemical composition is continuing to evolve as a result of nuclear fission and transmutation;
- Discovering the controlling factors that are key to materials performance and including them in models, which would reduce the likelihood of technical surprises;
- Discovering the controlling factors that are key to materials performance and including them in models, which, again, would reduce the likelihood of technical surprises;
- Transcending ideal materials systems to engineering materials and components; and
- Incorporating error assessments within each modeling scale and propagating the error through the scales to determine the appropriate confidence bounds on performance predictions.

If materials scientists can successfully meet these challenges, they will create nuclear materials performance models that can predict the properties, performance, and lifetime of nuclear fuels, cladding, and components in a variety of nuclear reactor types. They will be able to describe events throughout the entire reactor lifecycle and provide a scientific basis for the computationally based design of new, advanced materials.

The High-Dose Extreme

Industry needs models that predict the time dependence of microstructural and fission product evolution in structural materials and fuels. The most challenging extreme environment to study is that of high irradiation dose. Models developed to address this extreme are difficult to validate because of the inability to reach these doses using existing neutron-irradiation facilities in reasonable amounts of time and at modest costs. Furthermore, reactor facilities are problematic experimental venues for combining the various aspects of the extreme environments into a quantitative in situ study of material behavior.

Over the last several decades, the capability to study the high-dose regime and to conduct experiments that combine several components of the extreme environment to understand their synergistic effects has been demonstrated at ion-beam facilities around the world. Understanding radiation damage using ion irradiation is not a new idea. It has a long history of significant contributions spanning several decades. In fact, much of our understanding of material behavior under irradiation comes from well-controlled ion-irradiation experiments.

However, a key challenge in this effort is the scaling, or extension, of ion irradiation experiments and data to actual in-service conditions. Consequently a scientifically defendable argument for the applicability of models developed using accelerated experiments to neutron irradiation environments is critically needed. This should include rate scaling, effects of recoil energy spectra, and the ability to extrapolate to dose regimes not explored by neutrons. Where neutron irradiation experiments are not feasible, either because the appropriate spectrum is unavailable or because long irradiation times would be required, ion-beam experiments should be coupled with theory, simulation, and modeling and integrated using uncertainty quantification to effect advanced irradiation effects scaling. Advanced irradiation-effects scaling is identified as a priority research direction in the Science for Energy Technology workshop report (Crabtree and Malozemoff 2010). By definition, models developed for materials under neutron irradiation conditions at the extreme of high irradiation dose cannot be validated because little or no neutron irradiation data exists. Consequently, uncertainty quantification will be particularly important in quantifying uncertainties into dose regimes that have not been explored using neutrons.

Modeling Challenges for the Coupled Extremes of Irradiation, Corrosion, and Stress[2]

Advanced nuclear energy systems will require materials that can perform in aggressive environments for extended lifetimes under conditions that are close to safe operating limits. Some examples of such materials are: materials for high temperature gas-cooled reactor systems; high temperature, liquid metal or supercritical fluid systems; and advanced light-water reactor systems. In this context, a range of degradation mechanisms

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[2] Excerpted, with permission, from Moniz, Ernest, and Robert Rosner. 2009. Scientific Grand Challenges: Science Based Nuclear Energy Systems Enabled by Advanced Modeling and Simulation at the Extreme Scale. In *ASCR Program Documents*. Crystal City, VA: Office of Science, Advanced Scientific Computer Research and the Office of Nuclear Energy.

exist from general surface dissolution to localized corrosion, pitting, stress corrosion cracking, hydrogen embrittlement, liquid metal attack, oxidation, carburization, decarburization, etc. Such degradation occurs in the presence of intense radiation, high temperature, and mechanical stress, and includes a common underlying element, the interaction of an interface between a base material, often a metal alloy, and a gaseous or liquid environment. Figure 2 illustrates such an interaction between a metal and its environmental surroundings that results in irradiation assisted stress corrosion cracking (IASCC). In this process, cracking occurs only under the action of an aggressive environment *and* irradiation. The role of irradiation is important not only in how it changes the metal but also the water. Stress and high temperature provide additional driving forces for cracking. The evolution of thermal, micro-structural and stress environments under irradiation further aggravate the process by accelerating diffusion, creating new phases, inducing composition inhomogeneities, etc, that all play a role in the IASCC process. Thus, at the molecular scale, stress corrosion phenomena have their origins in the action of aggressive elements and anions in the interface layer with their environmental surroundings.

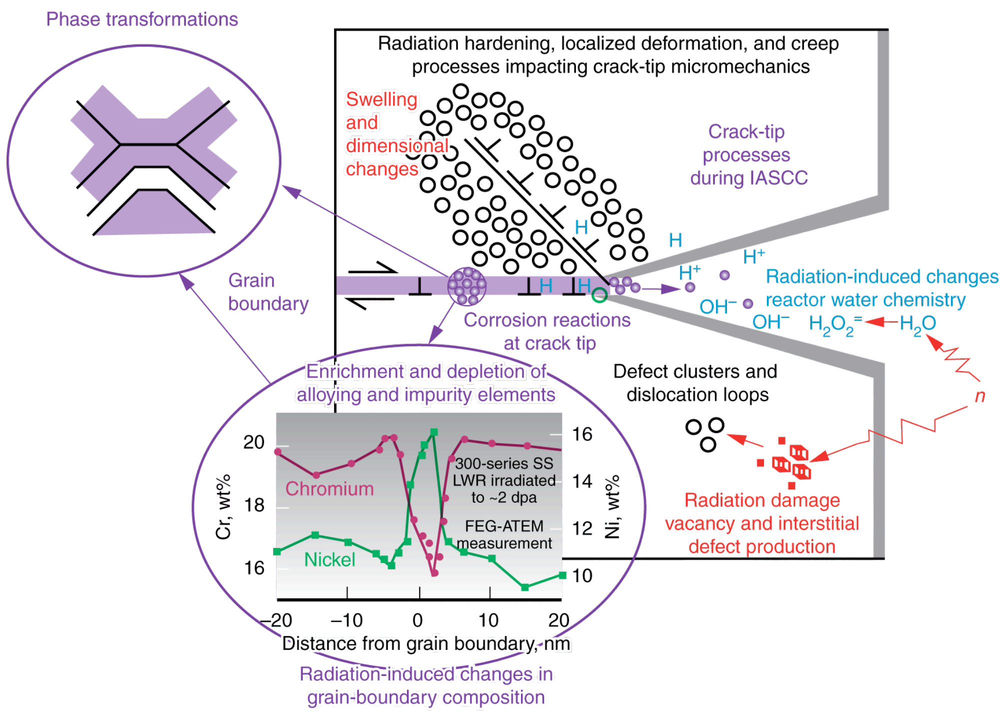

Figure 2 The phenomenon of irradiation assisted stress corrosion cracking embodies all of the elements of the extreme environment; radiation, stress, high temperature and corrosion, and presents a challenging modeling and experimental problem for predicting materials performance in reactor systems. (Source: Bruemmer, S. M., E. P. Simonen, P. M. Scott, P. L. Andresen, G. S. Was, and J. L. Nelson. 1999. Radiation-induced material changes and susceptibility to intergranular failure of light-water-reactor core internals. Journal of Nuclear Materials 274 (3):299-314.)

Modeling challenges include insufficient understanding of microstructure and microchemistry evolution of both fuel and clad and their interaction; modeling a discrete event, such as failure, especially given that multiple failure modes are likely to exist; and extending the models to fast reactor and TRISO fuel. Andresen has also detailed the essential elements of a successful SCC modeling program that includes treatment of the continuum in material, environment, and stress; treatment of time dependent crack growth to encompass the continuum from static, to slow strain rate, to cyclic loading; unified approach for crack, initiation and growth, which requires understanding of short crack behavior, fracture mechanics and crack chemistry similitude for relevance to varying component geometries and loading conditions; calculational approaches for complex service conditions which require accounting for the time and through-thickness variations in properties, and the use of distributions in properties as well as probabilistic approaches; integrated predictive modeling and on-line monitoring of system behavior extensibility into related cracking systems. (Andresen and Ford 1994)

Successfully applying advanced modeling and simulation techniques to the problem of environmental cracking is a formidable challenge, even in the absence of radiation damage. (Vashishta et al. 2007, 2008) Such an application is made difficult by the fundamental nature of chemo-mechanical phenomena that require that chemical reactivity and mechanical deformation be considered of equal importance. (Staehle 2005) Progress in applying such techniques to environmental cracking would very likely have a broad impact on the science and technology of materials performance.

Overview of the Initiative

The initiative aims to bring the methodology that is being successfully applied in NNSA's Stockpile Stewardship Program to accelerate the development of advanced nuclear energy materials. The focus is on predicting the behavior of cladding and actinide fuel in the extremes of high irradiation dose, transmutants and fission products, high temperatures, high stresses, and corrosive media. The initiative will develop time- and length-scale transcending models that predict material properties, supported by data from well-controlled ion-beam and neutron-irradiation experiments. The initiative will also use the methodology to design new radiation-tolerant materials.

This initiative seeks to build a scientifically defendable argument for the applicability of models developed using accelerated experiments to neutron irradiation environments. This should include rate scaling, effects of recoil energy spectra, and the ability to extrapolate to dose regimes not explored by neutrons. This initiative is distinguished by its use of uncertainty quantification to effectively integrate theory, simulation, and modeling with accelerated experimental capabilities. It brings together experts from universities, national laboratories, and industries to address critical issues in fuels and cladding.

In the following sections, we discuss the proposed methodology and five example problems that we call topical areas.

Approach

There are three essential elements of this initiative: uncertainty quantification; theory, simulation, and modeling; and accelerated experimentation. The relation of these three elements is shown schematically in Figure 3. The approach is analogous to that used in NNSA's Stockpile Stewardship Program, where a particular success was the plutonium-aging project that resulted in a savings to the taxpayer of $6–8B (Kintisch 2006).

QuesTek Innovations has adopted a similar approach and applied it to a computationally designed flight-qualified high-performance steel for landing gear applications. Ferrium S53 was developed with only five prototypes over a two-year period resulting in a development cost savings of approximately $50M (Anonymous 2003).

Models underpinned by UQ provide the bridge that links our high dose-rate experiments with reactor irradiations conditions

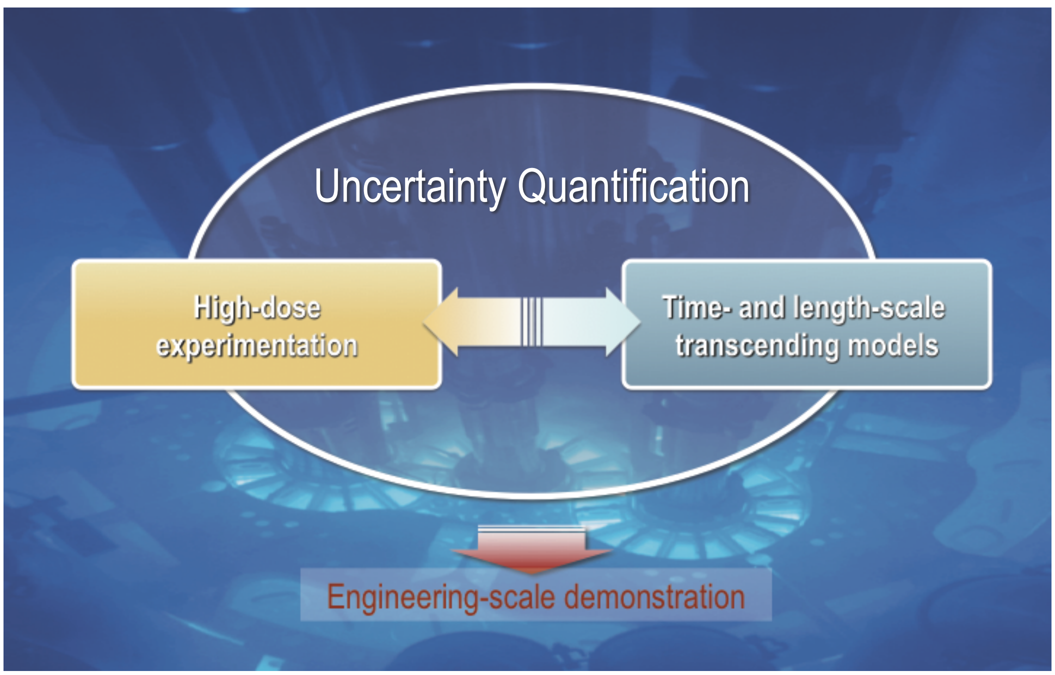

Figure 3. Schematic representation of the three essential elements of this initiative.

We seek to apply both of these successes to the development of advanced nuclear energy materials. In the following, we briefly describe the three elements, which are treated in more detail in Sections 2–4.

Why the Three Elements Are Required

It takes a long time to develop a new material or investigate the properties of materials that undergo low-dose-rate irradiation, such as the pressure vessel or core internals including the fuel and clad. Experiments using neutron irradiation can take up to 7 years, including the irradiation time, the radioactive cool-down time, and the post-irradiation examination. Incorporating the effects of high temperature, stresses and a corrosive environment along with irradiation make the problem multidimensional and extremely complicated. Translating that into a program to satisfy a license requirement for a new material or new fuel design can lead to a multi-decadal process. Such a timescale is unacceptable, and yet, it is the present-day norm.

One pathway to accelerate this process is to carry out accelerated experiments either inside the reactor core (in the case of the low-dose-rate regimes for the pressure vessel) or using external radiation sources such as ion beams (in the case of core internals that would see high neutron doses over their lifetimes). Decades of acceleration in dose rate can be achieved in days with ion beams.

Using ion beams, one can investigate a large phase space (in terms of external forcing functions) for irradiation in terms of both microstructure and macroscale properties. The phase space includes temperature, ion type, ion dose rate, ion energy, and total dose. It also includes the ability to apply in situ mechanical loading, chemical environments, and coolant fluids.

Probing that phase space, in some special cases, allows the creation of microstructures and material properties that follow the same path that would be found in a particular nuclear reactor irradiation experiment. However, microstructures and mechanical properties or other physical properties that deviate from neutron irradiations significantly will also be observed. The challenge is to employ that phase space to build a science-based understanding of material degradation and performance, specifically, to establish a scientific basis for the key mechanisms of materials performance.

The best approach to quantifying that scientific understanding is to build a model that captures all of the relevant physics, which is where modeling and simulation come into play. One seeks to understand the ion-beam forcing function and the material response to that forcing function. With that understanding, if a different boundary condition was applied (in terms of say temperature, ion type, and dose rate), it is reasonable to expect to be able to predict the material response. That is, one could have high enough confidence that, with this robust model, interpolation and reproduction of an experimental result is possible.

The question then becomes how does one extrapolate, given a model, to high-dose neutron-irradiation environments. Compared with ion irradiations, neutron dose rates are much, much lower (~10^2–10^3 lower). In addition, physical mechanisms, such as transmutation and chemistry changes, occur simultaneously with the ion bombardment and displacements in the material. With a robust model, the boundary conditions can be altered, while not perturbing any of the model internals, and an extrapolation can be made to project material performance to the neutron-irradiation environment. This approach has been termed advanced irradiation effects scaling. To make that extrapolation, researchers must first quantify the quality or accuracy of the extrapolation. This forward extrapolation and the qualification of the quality of the predicted extrapolation is where uncertainty quantification becomes important.

UQ also enables the inverse question to be tackled: given that the extrapolated response has a particular uncertainty associated with it, is it possible to ascribe or associate that uncertainty with one of the material model parameters? If so, is it possible to improve the model parameter using the ion environment so as to decrease the uncertainty in the neutron environment? UQ not only provides the forward extrapolation error estimation but also provides a tool for probing what type of conditions or experiments are necessary to achieve the highest quality extrapolated prediction in the neutron environment.

From this argument, an obvious question arises: is it possible to perform this type of program without all three of these components (UQ; accelerated experiments; and theory, simulation, and modeling)? For instance, is it possible to omit UQ and modeling and only do ion-irradiation experiments? Ion beams have been used to determine the exact temperature, dose rate, and ion type that would mimic the radiation damage seen in a neutron environment. However, this approach places intense pressure on being able to find the phase space of the material where that microstructure develops and then to re-create that phase space exactly so that one can extrapolate forward. In some cases, finding such conditions is impossible (Singh et al. 2000). In the methodology advanced for this initiative, if one combines modeling, simulation, and UQ, it is not necessary to exactly re-create the microstructures found in a neutron environment. Instead, one can create many different types of microstructures and use that entire knowledge base to extrapolate forward. The focus is on key mechanisms or developing a mechanistic understanding.

Similarly, if a program is built that only employs accelerated experimentation and modeling, the questions become, how good is the projection that has been made if it is not quantified in terms of a formal uncertainty quantification process, and what value would it have toward pursuit of licensing or any type of formal process for inserting materials into reactor environments.

Another scenario could be to eliminate accelerated experimentation and rely solely on a multiscale modeling campaign where one tries to bridge from understanding electronic structure to evaluating component performance. In this scenario, an implicit assumption is that models have the ability to accurately model all the physical processes with atomistic potentials. It is hard to baseline the model's performance or change any of its parameters, given experimental understanding, because no experimental data is available. This approach places enormous pressure on the modeling component to be more quantitative than it is in practice.

The realization is that all three elements—UQ; accelerated experiments; and theory, simulation, and modeling—must be brought together in a cohesive fashion to achieve the goals for this initiative.

Uncertainty Quantification

Uncertainty quantification is a decision-support methodology for complex technical decisions centering on performance thresholds and associated margins (Pilch, Trucano, and Helton 2006). It enables close integration of experiment with simulations and provides a management tool for prioritizing research. UQ has been applied with success under the NNSA's Stockpile Stewardship Program and is being studied for integration into the annual stockpile certification process.

Rigorous uncertainty quantification is critical in building credible simulation capabilities. It helps researchers answer questions such as how to evaluate whether theory, modeling, and experiment agree with each other; how to get them to agree with each other; and how to evaluate a model's predictive capability? It embodies five areas:

- Uncertainty analysis: What impact do parameter or model uncertainties have on model outputs?
- Sensitivity analysis: Which parameters contribute most to the output uncertainties?
- Inverse UQ: How can experimental data be used to better characterize parameter uncertainties?
- Calibration: How can experimental data be used to find the best parameter values?
- Risk analysis: In view of uncertainty, how do we quantify risk?

UQ is a tool that provides for management of research by focusing effort on the most important problems, identifying where experiment can provide input, and determining where additional theory, simulation, and modeling are required. UQ as applied in the Stockpile Stewardship Program has been reviewed by the National Research Council of the National Academy of Sciences (Adams et al. 2009). UQ has been the subject of several workshops: one related to grand challenges in national security (Higdon et al. 2009) and another on science-based nuclear energy systems (Adams et al. 2009).

UQ is treated in Section 2 by Richard Klein.

Accelerated Experimentation

The most challenging component of extreme conditions to study is high irradiation dose. Accelerated aging can and has been successfully performed with test reactors in the US and overseas. However, the ability to study the effects of high doses in existing test reactors is limited because the time and costs required to attain such doses are high. Furthermore, in reactor irradiations, it is challenging or impossible to separate the various aspects of the extreme environments for quantitative, in situ study of materials behavior.

Over the last several decades, the ability to study high irradiation doses as well as conduct experiments that combine several other components of extreme environments to understand their synergistic effects has been demonstrated around the world at ion-beam facilities. Ion irradiation has been used in studies of radiation damage for several decades, contributing much to the current understanding of material behavior under irradiation. In addition, Crabtree and Malozemoff (2010) have identified the need for advanced irradiation effects scaling.

Ion-beam facilities can be very flexible experimental platforms. In contrast with neutron irradiations, these experiments can be done at relatively low cost, with minimal sample activation, and in a greatly accelerated time frame. In fact, developing predictive models and new materials could be accelerated by 10 to100 times versus a reactor-based program, were the use of ion beams a possibility.

Yet, ion irradiation is not a substitute for neutron irradiation. Although these two methods share much in common, they result in distinct differences in the impact of particle type and dose rate on the irradiated microstructure. However, if ion irradiation is used to gain an understanding of how energetic particle–solid interaction links to the resulting microstructure and microchemistry, then models can be tailored for different particle types, fluxes, doses, and temperatures. That is, we posit that models underpinned and validated by ion-beam experiments and benchmarked against archival neutron-irradiation data will provide the required understanding and predictive capability to overcome the challenges posed by extreme environments.

Developing predictive models requires a strong theory and simulation effort. This initiative should lead to the development of predictive models with ion-irradiation experiments serving to identify unit mechanisms, validating the models, and providing material property data. Only recently have computational techniques and approaches evolved to the point where the effects of extreme environments on concentrated alloys and fuels with complex, realistic microstructures can be modeled. The coincident rise in the capability of ion irradiation as a valuable tool and the maturing of modeling and simulation presents an opportunity for advancement that has not been possible until now.

Accelerated experimentation using ion beams is treated in Section 4 by Gary Was.

Time- and Length-Scale Transcending Models

Multiscale material modeling has been a focus at Lawrence Livermore National Laboratory (LLNL) since the 1990s. This work is driven by the mission-based need to develop physics-based models for use in continuum code simulations. The potential to develop predictive models built from fundamental building blocks is a very attractive paradigm. In practice, the atoms-to-continuum approach is difficult to implement because the broadly diverse and interesting technical issues at the lowest length scale tend to consume resources, leaving little for work on the continuum scale. Therefore, the approach of this initiative is to start with the highest level continuum model that represents the physics of interest and work toward the lower length scales, obtaining parameters experimentally when possible and appealing to more physics-based models when necessary. This process naturally produces models that are treated at the level of specificity required to meet the need and no more.

Multiscale models, funded by NNSA through the Advanced Simulation and Computing (ASC) Program, predict material strength in extremes and across orders of magnitude in strain rate. The box on p. 16 provides two examples of successful multiscale models. An important breakthrough that enables this initiative to go forward is progress made in time-transcending modeling. An example of this class of model is given in the box on p. 17.

As mentioned above, models are most useful when they used in continuum code simulations adapted for practical application. It is for this purpose that we selected the FRAPCON code (Lanning, Beyer, and Painter 1997; Berna et al. 1997; Bernd et al. 1997) and its associated MATPRO material model database (Allison et al. 1997) for developing models of LWR materials.

Time- and length-scale bridging modeling is treated in Section 3 by Vasily Bulatov.

Model Validation and Data Sources

Models will be validated against data from ion irradiation, neutron irradiation, and archival neutron-irradiated samples. Both ion and neutron irradiations can be performed under controlled temperature. The target temperatures and the variability in temperature possible in neutron and ion experiments are similar. Ion-beam experiments, by their nature, are single-energy irradiations, while neutron experiments can use filters and shielding to focus on portions of the neutron spectrum. Both ion beams and reactors can conduct irradiations over a range of times from minutes to hours. The higher allowable dose rates of ion beams allow an experiment to reach a higher dose in a shorter period of time; however, the irradiated volumes are much smaller, and care must taken to understand the effect of dose rate on the ensuing microstructure. The Advanced Test Reactor (ATR) National Scientific User Facility (NSUF) is the natural partner to this initiative because, as a DOE NE facility, it has a mission to help contribute to the understanding of materials and fuels in nuclear systems. The ATR NSUF provides the ability to support four critical aspects of this initiative, offering access to: (1) neutron-irradiation facilities to carry out tailored energy and integrated neutron-irradiation experiments, (2) libraries of neutron-irradiated samples and unirradiated samples of the same materials, (3) modern analytical capability to analyze samples with residual radioactivity from irradiation, and (4) ion-irradiation facilities. The type of experiment (whether neutron or ion) will be chosen to answer specific scientific questions or to validate specific modeling calculations.

Multiscale Modeling of Tantalum and Vanadium: A Success Story

NNSA's ASC Program has funded a multiscale modeling effort in support of the Stockpile Stewardship Program to predict the mechanical properties of materials under the extreme conditions of high pressures, strain rates, and temperatures. The challenge of this effort is to make such predictions with simulation alone and in the absence of experimental data. The experimental data obtained under such conditions is integral to model development. However, although the quality of the available data is sufficient for validating models, it is not high enough for developing them. Within this context, a multiscale materials modeling effort is engaged in developing constitutive models of material strength and strain hardening capable of being applied in engineering finite-element analyses from information gained from a hierarchical assembly of material physics simulations. In this incarnation, electronic structure simulations are used to determine equations of state, phase and crystal symmetry, elastic constants, and other pertinent information for building high-quality molecular potentials. Molecular dynamics and statics simulations calculate dislocation core energies and selected dislocation mobilities (stress–velocity relationships), as dislocations are known to be the relevant defects for plastic relaxation. Large-scale dislocation dynamics simulations then integrate the behaviors of individual dislocations to the behavior of complex networks of defects. The outputs of dislocation dynamics provide the initial yield strength and strain hardening characteristics as well as the details of the corresponding dislocation microstructure evolution, which are used to inform physics-based strength model. Finally, continuum single- and polycrystalline plasticity simulations homogenize the dislocation dynamics outputs into an effective medium. The resulting models are then implemented into finite-element codes to simulate the growth of engineered perturbations during a laser shock experiment, as shown in Figure 4. The growth of these perturbations is sensitive to material strength and strain hardening at high pressures and strain rates. The model built with simulation input alone and no experimental data performed better than classical models that were fit to data in other regimes and used to extrapolate to the conditions of materials under laser shock conditions. Furthermore, even without calibration, the simulation-only model performed comparably to the classical model under the conditions at which the classical model had been calibrated.

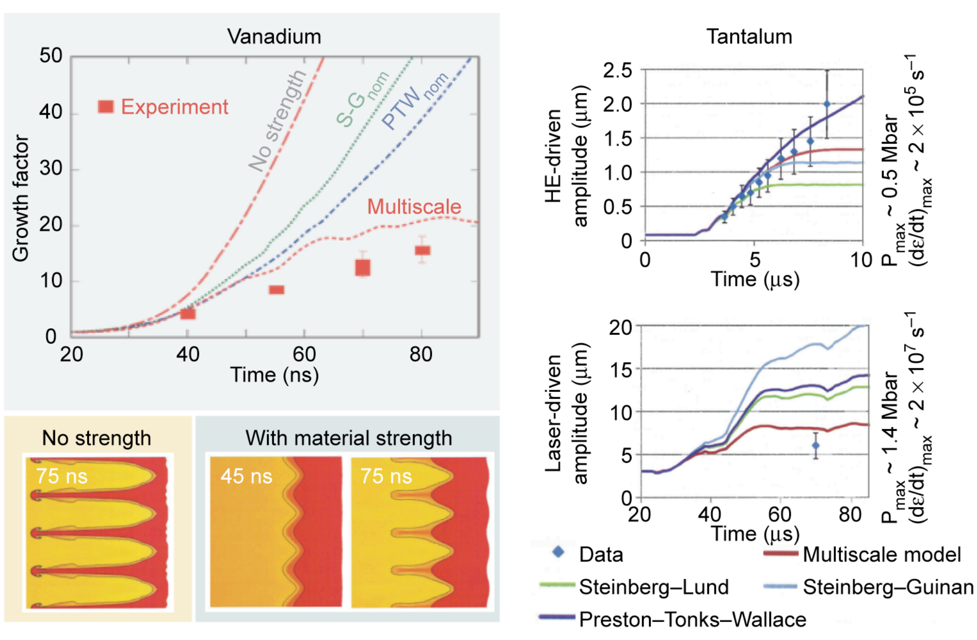

Figure 4. Example results from multiscale models compared with those from classical approaches.

Time-Transcending Models

The premise of the proposed initiative is that theory, modeling, and simulations can provide a reliable connection between material behavior observed over hours and days of violent (high-dose-rate) ion-beam irradiations in accelerated tests and the expected performance of the same material under much lower dose rates but over much longer working life in a reactor. In Figure 5, the top-left image is a computer-simulated damage microstructure in α-iron after 8 hours of electron irradiation under a dose rate of 10^{-4} dpa/s and at 200°C. Red spheres indicate vacancy clusters, and blue is interstitial clusters. The top-right image shows results when the same damage dose was simulated at the same temperature but under a much lower does rate of 10^{-8} dpa/s. The accumulated damage under these conditions is much lower, showing only very few vacancy clusters in the same material volume. This α-Fe model was then used to determine if the accelerated damage (accumulated under high dose rate in an accelerated test) can be adjusted to resemble damage received under prolonged irradiation. The two images in the bottom row show damage accumulated after the same dose of irradiation under the high (left) and low (right) dose rates. Although differing in details, the resulting microstructures are substantially similar. The approximate correspondence was achieved here by using an elevated temperature to accelerate the rate of damage annealing under the high-dose-rate irradiation. Competition of multiple kinetic mechanisms of damage accumulation makes it impossible to scale the predicted damage exactly, even with this relatively simple and well-studied material (α-Fe). At the same time, accurate and computationally efficient simulations can be used to explore and identify experimental conditions in which accelerated material tests would be most informative for model validation, e.g., most sensitive to a particular mechanism or model parameter. Furthermore, the same simulations can be applied to fine-tune the models for conditions typical of real reactors. The approximate scaling demonstrated here provides exactly the right kind of connection between simulations and experiments. Furthermore, once the accuracy of the kinetic model is established, simulations can be used to extrapolate from accelerated material tests into relevant but inaccessible conditions of nuclear reactors without relying on any approximate scaling. The reliability of such *computational extrapolations* can be quantified by using these kinetic simulations to assess the uncertainties in the computational predictions of accumulated damage, given the uncertainties in the model parameters.

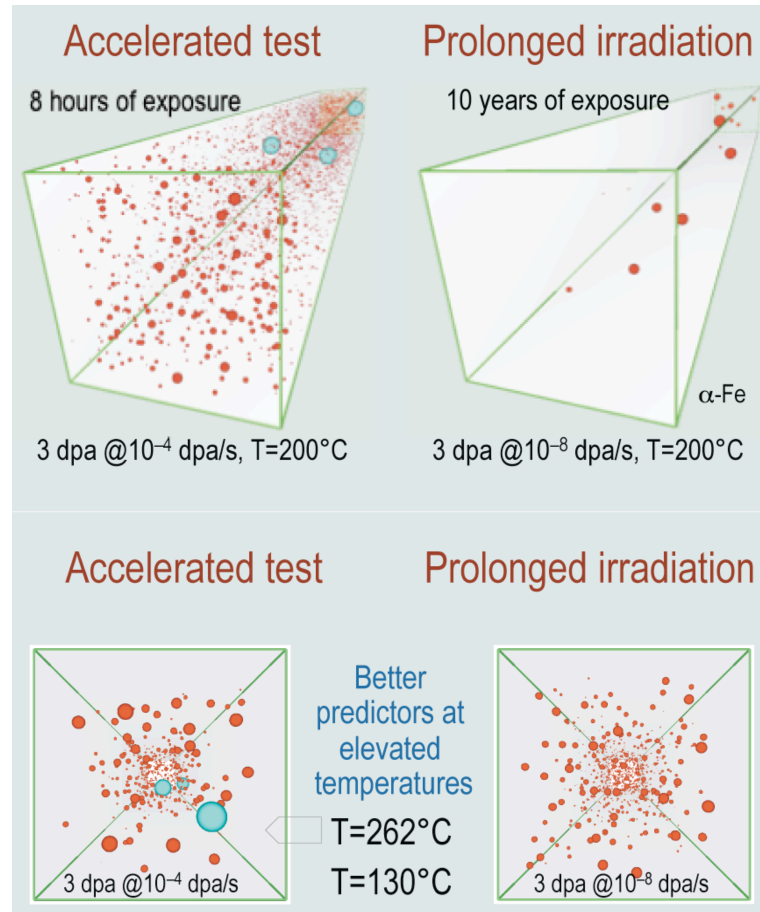

Figure 5. Results from computer simulations predict how different irradiation conditions will affect the microstructure of α-iron.

Timeliness

In the last decade, significant advances have been made in theory, simulation, and modeling. Successful examples abound, including density functional theory (DFT), accelerated molecular dynamics (MD), dislocation dynamics (DD), first passage kinetic Monte Carlo (KMC), successful multiscale model, terascale computing, energy-filtered high-resolution electron microscope (HREM) imaging, atom probe, aberration-corrected HREM, in situ experimentation, micromechanical testing, multi-ion-beam capabilities in Japan and France to probe synergistic effects, transmission electron microscope (TEM) ion-beam interfaces, and uncertainty quantification for plutonium aging and stockpile stewardship. Together, these advances provide new impetus for using ion beams to probe the extreme of high irradiation dose.

During the next 20 years, critical decisions will need to be made regarding the future of neutron-irradiation facilities in the US (e.g., see Section 10: Naval Reactors). With the available reactor test platforms becoming heavily subscribed and construction of new facilities uncertain, further systematic advances in our fundamental understanding of irradiation effects can be strongly supported with ion-based studies coupled with advanced simulation and modeling. The use of UQ to integrate accelerated experiments with advanced simulation and modeling can accelerate radiation damage science, thus shortening the design cycle and reducing costs. Ion beams coupled with advanced simulation and modeling can provide actionable information on materials performance and thus enhance the value of existing neutron-irradiation facilities. Now is the time to develop validated techniques for applying ion-beam capabilities to evaluate the behavior of reactor fuels and materials outside of actual in-service environments.

Relation to the NE programs

The proposed initiative has synergy with a number of existing NE programs including Advanced Modeling and Simulation, Fuel Cycle Research and Development, Gen-IV, and Light Water Reactor Sustainability. The methodology proposed in this initiative could complement existing efforts, contributing through the development of new models and materials.

Advanced Modeling and Simulation. NEAMS is focused on building predictive simulation capabilities of the performance and safety of integrated systems (reactors, fuels, safeguarded separation processes, waste and repositories). To do that, NEAMS is relying on fundamental material behaviors as much as possible. The proposed initiative discusses the challenges and possible opportunities to improve material models using uncertainty quantification to integrate theory, simulation, and modeling with accelerated experimentation. The improved models could then be adopted by NEAMS and used to create improved integrated system level simulation capabilities.

Fuel Cycle Research & Development. The advanced fuels campaign has as part of its mission and objectives a focus on high burnup LWR fuels and deep burn fuels. There is a need to assess fuel performance by accelerated probing of unit mechanisms operating in fuels under reactor conditions to deliver in a timely fashion critical information on fuel properties (second phase precipitation, enhanced diffusion, microstructural changes, etc.)

and performance. A science-based approach, such as that proposed in this initiative and which is part of the current NEAMS vision and Fuel's Integrated Performance and Safety Codes (IPSC) and Verification & Validation and Uncertainty Quantification (VU) program elements, would accelerate and guide the classical empirical (prototype-based) fuel qualification process that has been lengthy, expensive and does not always provide optimum performance. Although applications of UQ are embedded in NEAMS, UQ applied to nuclear energy materials is an integral part (the foundation) within the proposed initiative. The initiative proposes to apply UQ to effectively integrate theory, simulation, and modeling with high-dose experimental capabilities to accelerate the design and development of fuels and cladding. This integrated approach complements the existing program elements.

Advanced Concepts. Concepts for advanced nuclear reactors emphasize more efficient use of the nuclear fuel, resulting in higher irradiation doses to both fuel and structural materials. This implies the need for new radiation tolerant materials. Combining accelerated experiments as proposed in this initiative with an advanced modeling capability and integrated using UQ would allow researchers to better understand material's behavior over time and could contribute to decreasing the time needed to develop breakthrough materials required for new systems (see Stakeholder discussion).

Light Water Reactor Sustainability. Extending nuclear reactor operations to 60 years and beyond requires more fundamental knowledge of materials degradation phenomena. Uncertainty quantification, used to integrate theory, simulation, and modeling with accelerated experimentation offers a methodology to develop models and materials that could contribute to this knowledge and therefore to the technical case for life extension.

In addition, as stated in the Stakeholder discussion, "The time and cost associated with developing and licensing new materials is a considerable barrier to introducing new nuclear fuel technologies. Establishing techniques to more efficiently identify new materials and then to collect the performance data needed for licensing new technologies seems appropriate for DOE funding, and a task for which the Department's research community is well suited."

Goals and Five Topical Areas

The scope of this initiative is responsive to a request from NE to focus on fuels and cladding. Related work may already be ongoing in DOE by other organizations. The topical areas below are designed to demonstrate the added contribution brought by this initiative. In the following, five specific problems to be undertaken are described. These are graded in difficulty and cost. The problems can be sequenced to demonstrate incremental levels of success.

This initiative has two overarching goals and five closely related topical areas. The topical areas listed below were the proposed TAs presented at the May 11 workshop. In Sections 5–9, we describe how we used input from the workshop participants to sharpen and refine these five areas.

Initiative Goals

The two overarching goals for this initiative are to (1) develop time- and length-scale transcending models that predict material properties using uncertainty quantification to effectively integrate theory, simulation, and modeling with high-dose experimental capabilities, a methodology that has been highly successful in the Stockpile Stewardship Program; and (2) design and develop new radiation-tolerant materials using the knowledge gained and methodologies created to shorten the development and qualification time and reduce cost.

Pre-workshop Topical Areas

TA1: Model Development. Develop a validated multiscale model that predicts the yield point and hardening rate of a neutron-irradiated alloy using UQ to integrate theory, simulation, and modeling with accelerated experimentation.

TA2: Chemistry and Property Evolution of Fuels. Develop a multi-physics model to predict site redistribution and fission-gas release, and their consequences on the thermal conductivity of metallic U-Zr and U-Pu-Zr fuels irradiated to 20 at.% burn-up with operating temperature in the central region of the fuel pin up to 750°C using UQ to integrate theory, simulation, and modeling with accelerated experimentation.

TA3: Advanced Clad Materials Development. Predict yield point and hardening rate, including irradiation creep effects and irradiation growth of zirconium alloy cladding irradiated to 100 MWd/MTU under the following environmental conditions: operating temperatures of 290–360°C and a loss-of-coolant accident (LOCA) conditions up to 1000°C using UQ to integrate theory, simulation, and modeling with accelerated experimentation.

TA4: Fuel–Clad Interaction Leading to Fuel Failure. Model high-burn-up, high-power-density failure modes of zirconium alloy cladding in contact with oxide fuel under the following environmental conditions: operating temperatures of 290–360°C and LOCA conditions up to 1000°C, water chemistries including Li/B-containing pressurized water reactor (PWR) coolant and boiling water reactor (BWR) normal and hydrogen water chemistries, temperature, and stress using UQ to integrate theory, simulation, and modeling with accelerated experimentation.

TA5: Radiation-Tolerant Materials. Design and develop materials that mitigate or even eliminate the effects of the extreme irradiation environments using UQ to integrate theory, simulation, and modeling with accelerated experimentation. Design and develop materials that exhibit smaller, more predictable variability in properties and performance using UQ to integrate theory, simulation, and modeling with accelerated experimentation.

Summary

This initiative seeks to introduce a three-pronged model development approach to the NE Program. Its distinguishing feature is the use of uncertainty quantification to effectively integrate theory, simulation, and modeling with high-dose experimental capabilities. The initiative brings together experts from universities, national laboratories, and industries to address critical issues in fuels and cladding. We will develop time- and length-scale transcending models that predict material properties, supported by a key understanding of the unit processes that drive the microstructure and by data from well-controlled ion-beam and corresponding neutron-based experiments. Our focus is on predicting the behavior of cladding and actinide fuel in the extremes of high irradiation dose, transmutants and fission products, high temperatures, high stresses, and corrosive media.

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2. Uncertainty Quantification

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Uncertainty quantification studies all sources of error and uncertainty, including: systematic and stochastic measurement error; ignorance; limitations of theoretical models; limitations of numerical representations of those models; limitations on the accuracy and reliability of computations, approximations, and algorithms; and human error. More precisely, UQ is the end-to-end study of the reliability of scientific inferences that results in a quantitative assessment of that reliability and provides inventories of (1) the possible sources of error and uncertainty in the inferences and predictions, (2) the sources of error and uncertainty accounted for in the assessment, and (3) the assumptions on which the assessment is based.

Advances in computing over the past few decades—both in availability and power—have led to an explosion in computational models available for simulating a wide variety of complex physical (and social) systems. These complex models—which may involve millions of lines of code and require extreme computing resources—allow researchers to simulate physical processes in environments and conditions that are difficult or even impossible to access experimentally. As a result, they have led to numerous scientific discoveries and advances.

However, scientists' abilities to quantify uncertainties in these model-based predictions lag well behind their abilities to produce the computational models. This is largely because such simulation-based scientific investigations present a set of challenges that is not present in traditional investigations; for example:

- The amount of physical data (observational or experimental) is typically quite limited.
- The computational demands of the model limit the number of simulations that can be carried out.
- The computational models are not perfect representations of physical reality—they have inadequacies, approximations, missing physics, etc.
- The computational models typically include unknown parameters and boundary conditions that must be adjusted for the application at hand.
- Researchers often want to extrapolate such models to conditions where they have little or no physical observations to validate model output.

With UQ, researchers have tools to address several key issues. This approach will allow them to determine which model parameters contribute most to the output uncertainties, what impact parameter or model uncertainties have on model outputs, and how experimental data can be applied to better characterize parameter uncertainties or to find the best parameter values. It will also help them answer an important question related to predictive simulations: In view of uncertainty, how do we quantify risk? Used properly, UQ can be a unifying framework for tying together theory, simulation, and experiment.

Developing a powerful UQ capability will:

- Yield critical insight into scientific predictions of great impact in both national and international arenas (e.g., climate).
- Drive informed decisions on new designs of critical experiments in many scientific fields (e.g., inertial confinement fusion [ICF] capsule design, magnetic fusion energy, NE).
- Assign confidence bounds to outcomes (e.g., climate, science-based stockpile stewardship).
- Allow disciplines that cannot conduct experiments to directly test important predictions.

In the current state-of-the-art practice of UQ, we take the following approach:

- Assume that the discretization errors in partial differential equations (PDEs) are smaller than the uncertainties.
- Utilize an ensemble of models assuming that uncertainties are dominated by a small number (usually ~ 7–10) of parameters.
- Sample these parameters using standard techniques (e.g., Hybrid Latin Hypercube sampling, Monte Carlo).
- Compute the model with simulator (code) runs from a few to thousands of times (ensembles).
- Use subsets of sample ensembles to construct a surrogate emulator (e.g., response surface, meta-model) in N-dimensional space to produce multivariate adaptive regression splines, Gaussian process models, etc.
- Use the response surface to bootstrap results to regimes outside the experimentally validated regime and perform statistical analysis of the N-dimensional response surface.

Other approaches rely on bounding inequalities (e.g., concentration of measure; Lucas et al. 2008). Many of them can be cast in the framework of a Bayesian computational inference engine.

Several key grand challenges remain at the forefront of current research in UQ. Those that warrant significant advances include (1) dealing with the curse of dimensionality where the sample point density (ensemble runs of numerical simulations) decreases exponentially as the dimensionality of the uncertainty phase space increases, rapidly resulting in a computationally prohibitive obstacle to quantifying uncertainty in a high dimensional uncertainty space; (2) propagation of uncertainty and error in forward simulation; (3) aggregation of error and uncertainty; (4) quantifying uncertainty of rare outcomes that may have significant impact; (5) quantifying uncertainty in extrapolation to regimes for which experimental data is scarce or non-existent; and (6) exploring UQ at the extreme scale (e.g., exascale). To address many of these challenges, LLNL has created an advanced UQ science capability for predictive simulation. This effort involves 21 physicists, mathematicians, statisticians, and computer scientists working in three interrelated technical areas to build a UQ computational engine for exascale computing:

- **Error Estimation:** Discretization error estimation in multi-physics and multiscale algorithms and codes.
- **Curse of Dimensionality:** Research in non-intrusive techniques, such as dimension reduction, adaptive sample refinement, advanced response models, and topological characterization.
- **UQ Pipeline:** Workflow management with self-guiding, self-adaptation UQ data analysis and visualization.

The initial application areas addressed in the LLNL strategic initiative are science-based stockpile stewardship, climate prediction, and ICF. The methodologies developed under the LLNL effort can readily be applied to the proposed initiative for NE.

To address the curse of dimensionality, research efforts have begun in:

- **Adaptive Sample Refinement (ASR):** Development of a methodology to adaptively (sequentially) construct informative and efficient ensembles in UQ runs.
- **Dimension-Reduction/Variable-Selection (DR/VS):** Development of input/output techniques to reduce the numbers of dimensions and select the input variables.
- **Advanced Response-Model Methodology for UQ:** Research in robust high-dimensional response models to advance the response-model-based UQ methodology (for example, to better understand error processes, bound uncertainty, etc.).
- **Topological Techniques:** Research in the use of topological structures, such as contour trees, to analyze, compare, and visualize high-dimensional response functions. Contour trees represent all level sets of a scalar function as well as their nesting behavior. They provide a compact view of the function without resorting to traditional dimension reductions, and they preserve all critical points and important neighborhood information. They also represent an optimal data structure for extracting level sets. The main focus of this research effort is to develop new topological schemes to analyze response-model surfaces and UQ misfit functions (likelihood, posterior probability distribution function), to research methods that use topographical instability to guide the ASR process, and to develop intuitive representation of the global topological structure of a high-dimensional response surface.

The complexity of performing end-to-end UQ analysis requires the development of an advanced UQ computational pipeline. At LLNL, we are developing self-guiding, self-adapting technologies to allow the UQ pipeline to automatically steer the selection of ensemble simulations to cover the complex topology of the response domain most efficiently and perform end-to-end UQ. The advanced pipeline will determine what additional ensemble simulations are needed to sample the uncertainty phase space (self-guiding feature) and will embody a decision-making capability that chooses the best sampling and dimensional reduction strategies to employ for the UQ study of interest (self-adapting feature). A codex being developed will enable the self-guiding, self-adapting technologies in the UQ pipeline. The UQ pipeline framework is general and will be adapted to any simulation code such as FRAPCON or the relevant material simulation codes in NE to permit UQ analysis.

Non-intrusive UQ using an ensemble-based approach attempts to take into account all sources of input error associated with the simulations as well as error associated with the measurement observations. The categories of uncertainty or error include error in the observational measurements; prediction error or uncertainty associated with the statistical emulator (response surface or meta-model), both for current conditions where we have observations and for the new conditions where we might not have any observations; and uncertainty in the simulation code unexplained by the parametric uncertainty we have included for the models. This is frequently referred to as the *unknown unknown* (or as model error or model inadequacy). How can we get a quantitative understanding of the so-called unknown unknowns? The approach that UQ takes is to simultaneously account for (1) observational error, (2) emulation error, and (3) model inadequacy error. The reason being that in most cases one does not have the necessary volume of observations or simulations to characterize each of these terms separately. Hence, given the observations we have and a fixed number of simulations, we can aim to characterize the joint impact of all these sources of error of both UQ of the parameters in the models and the prediction of the state of the system for the new regime we are aiming for. By carrying out a large ensemble of simulations to develop response surfaces and then comparing those predictions with random sets of additional simulations not used to construct the response surface, we can both quantify and reduce the emulation error term. We might also produce a good characterization of the observation error term. The model error (unknown unknowns) is what remains: the difference between the simulations and the observations that we cannot explain by observation–emulation uncertainty. As we made assumptions about the statistical properties of the observation errors and the response model, we make a prior statistical assumption about the model error. By working simultaneously with all three sources of errors, we get a quantitative idea about the contribution of unknown unknowns (i.e., the model error term).

We have applied the ensemble of models approach to UQ in the areas of science-based stockpile stewardship, ICF, and most recently, climate atmospheric prediction. Using our UQ sensitivity approaches in the area of climate prediction, we have screened and ranked the most important contributors of uncertainty in key model outputs and narrowed down the parametric uncertainty in climate atmosphere models to a 15-dimensional uncertainty space after starting with a 100-dimensional uncertainty space. By using existing satellite measurements and the accompanying measurement error, we can further constrain the uncertainty in the atmospheric model parameters. We are attempting to use this reduced parametric uncertainty response model to perform statistical analysis and determine the uncertainty in a variety of metric outputs relevant to climate prediction.

UQ Approach for the Proposed NE Initiative

In this initiative, UQ is both a tool for prioritizing research and quantifying uncertainties when extrapolating into dose regimes that have not been explored using neutrons. Figure 6 gives a simplified flowchart illustrating how UQ could be applied to accelerate the development of nuclear energy materials. UQ requires a model. Since the goal is to insert models into continuum code simulations, the model should be the highest-level continuum code simulation that contains the physics relevant to the problem. (If no model exists, ion-beam data could be acquired as a function of dose, dose rate, ion energy, ion type, temperature, alloy composition, etc., filling in a large matrix of experimental conditions. The experimental input could then be used to models developed that describe this behavior.) Aleatoric and epistemic uncertainties would be estimated and used to compute which parameters contribute most to the output uncertainties and the impact that parameter or model uncertainties have on model outputs.

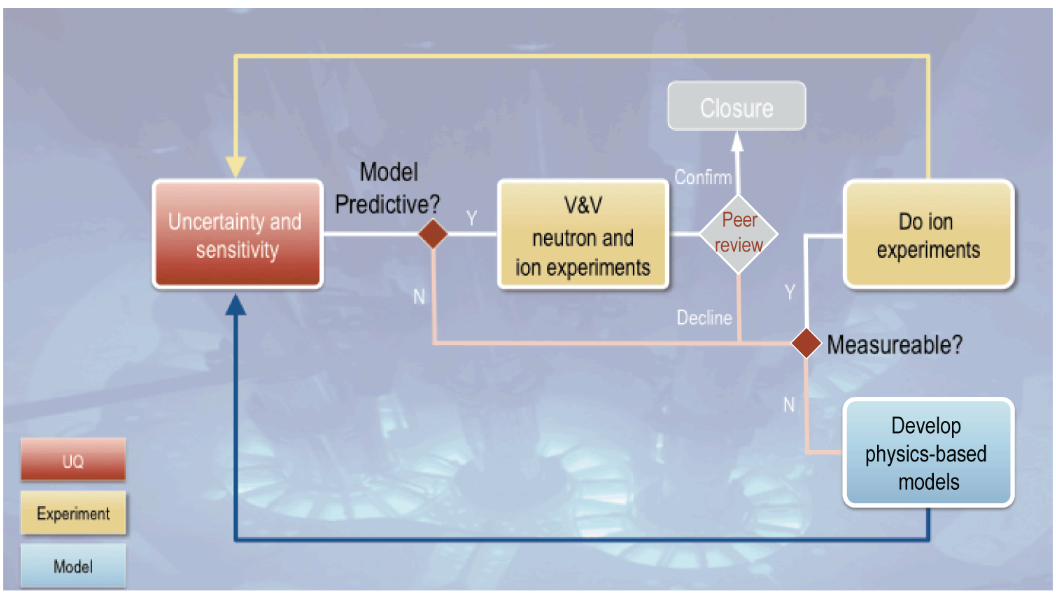

Figure 6. Schematic diagram illustrating how UQ will be applied in this initiative.

Using inverse UQ, we would prioritize parameters based on their combined uncertainty or sensitivity (Figure 7). If the uncertainties and sensitivities are outside acceptable bounds, the model is deemed to not be predictive, and better estimates for the prioritized parameters are sought. The model is used to help determine if the parameters of interest can be measured experimentally (calibration). If they can, experiments are conducted. If the experiments are not sensitive to the parameters, an appeal is made to more physics-based models.

In either case, the cycle begins again and continues until the uncertainties and the sensitivities are acceptable. The resulting model is then subjected to verification and validation, carrying out experiments at both ion- and neutron-irradiation rates, if possible.

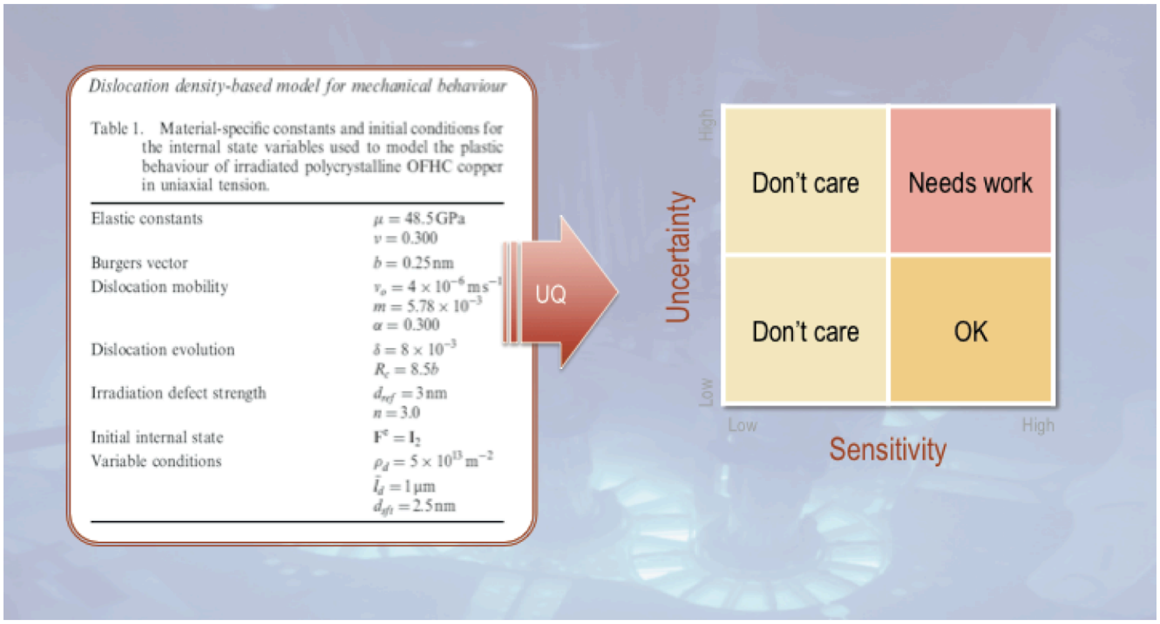

Figure 7. Using inverse UQ, parameters are prioritized for study. Those parameters that give rise to high uncertainty in the model output and with high sensitivity are given highest priority. (From Arsenlis, A., Wirth, B. D., and Rhee, M., 2004. Dislocation density-based constitutive model for the mechanical behavior of irradiated Cu. *Philos. Mag.* **84**(34), 3617–3635.

Because the methodology starts with a high-level continuum model, there is always the possibility of compensating errors in the analysis. The model and all associated experimental results will then be subjected to expert peer review. Once the expert community is convinced that the model is predictive, the problem is considered solved.

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3. Theory, Simulation, and Modeling

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Theory, simulation, and modeling (TSM) are viewed as an integral part and a substantial driver of the proposed initiative. Uncertainty quantification provides a logical and consistent framework for TSM development and dictates that: (1) construction of a multiscale multi-physics modeling hierarchy should proceed in the top-down direction, from engineering (macroscopic) scales to mesoscopic to atomistic; (2) first UQ cycles should be performed on existing models; and (3) development of subscale models is undertaken only when needed to reduce uncertainties in the larger-scale models.

The role of TSM in this initiative is to provide trustworthy predictions of expected material performance under reactor conditions (e.g., long-time exposure to low-rate neutron irradiation, transmutant gas accumulation, corrosion) from the behavior of the same material under short exposures to high-dose-rate ion beam irradiations. To serve in this important role as a computational bridge, the models of material degradation must be validated against the high-rate ion-beam test data and have sufficiently low uncertainties to allow reliable predictions on the in-reactor timescales.

The key technical challenge for TSM is to be able to transcend the length- and time-scale gaps between the initial damage production in the form of collision sub-cascades (10 ps, 10 nm) to the scales relevant for engineering calculations of reactor components (tens of microns, many years). Of the two scale gaps, timescale disparity is the more serious one: it spans over 20 decades. In addition time integration, unlike space, is inherently sequential and difficult, if not impossible, to accelerate through massively parallel computing. In materials simulations in general and in simulations of irradiated materials in particular, the requirement of model accuracy conflicts with simulation efficiency: simulations (or calculations) that are more faithful to the fundamental physics of material behavior invariably incur higher computational cost.

One can distinguish two logically, if not physically, separate TSM elements of material degradation under irradiation. The first is TSM of microstructure evolution and damage accumulation with no regard to the resulting mechanical properties. The second is TSM of mechanical property degradation for a given damage microstructure. Models and simulation algorithms for these two elements differ substantially, although unification of the two elements in a single modeling framework is possible and even desirable in the future.

Multiscale simulations of damage accumulations use input from atomistic simulations and experiments to identify and quantify diffusion-reaction mechanisms by which defects introduced in the lattice by the initial collision cascades (damage source) gradually evolve to produce the resulting damage microstructure. Several modeling approaches have been used to simulate damage accumulations: rate theory (RT, also known as cluster dynamics), object kinetic Monte Carlo (OKMC), phase fields, lattice gas models, and even atomistic simulations. Of these, RT is the workhorse method in use since the 1960s.

Recent improvements in the efficiency of the OKMC method now allows researchers to simulate damage accumulation on timescales of years and beyond while, at the same time, dispensing with the crude mean-field assumption used in the RT formulation. RT, OKMC, and other approaches for modeling damage accumulation are the focus of ongoing development efforts across the TSM community, and substantial progress is being reported in making these methods more accurate and computationally efficient. This progress bodes well for the proposed initiative—we will rely on the more computationally expedient models for initial UQ cycles but will switch to more physically based models if and when their computational efficiency becomes acceptable for uncertainty propagation.

Several models of mechanical strength degradation exist, some of which are included in the engineering multi-physics codes (e.g., FRAPCON). These can be used for UQ cycles. However, several workshop participants expressed concern at the validity of such models and their reliability for computational bridging from accelerated ion-beam tests to in-reactor conditions. The concerns were expressed in terms of "immature," "not physically based," etc. Alternatively, one can observe that little if any information on the evolving damage microstructure is included in most models of strength degradation currently in use. Several research groups in Europe, Japan, and the US are examining effects of irradiation-induced damage on crystal strength. Most of the ongoing efforts are focused on the interactions of a single dislocation with one or a few defects, whereas understanding of collective mesoscale phenomena (e.g., channel formation) is scarce or lacking. This problem is likely to be resolved in the near future with the availability of novel mesoscopic simulations methods, such as dislocation dynamics and phase fields, thus improving the TSM effort to evaluate damage effects on material strength (Figure 8). Based on the analysis of such mesoscopic simulations, one can envision development of continuum constitutive functions relating stress to strain rate and coupled with a small set of evolution equations for coarse-grained microstructural variables, such as dislocation and loops densities. A similar approach worked effectively in the parameter-free modeling of crystal plasticity in extreme deformation conditions within an ASC Program effort focused on materials dynamics.

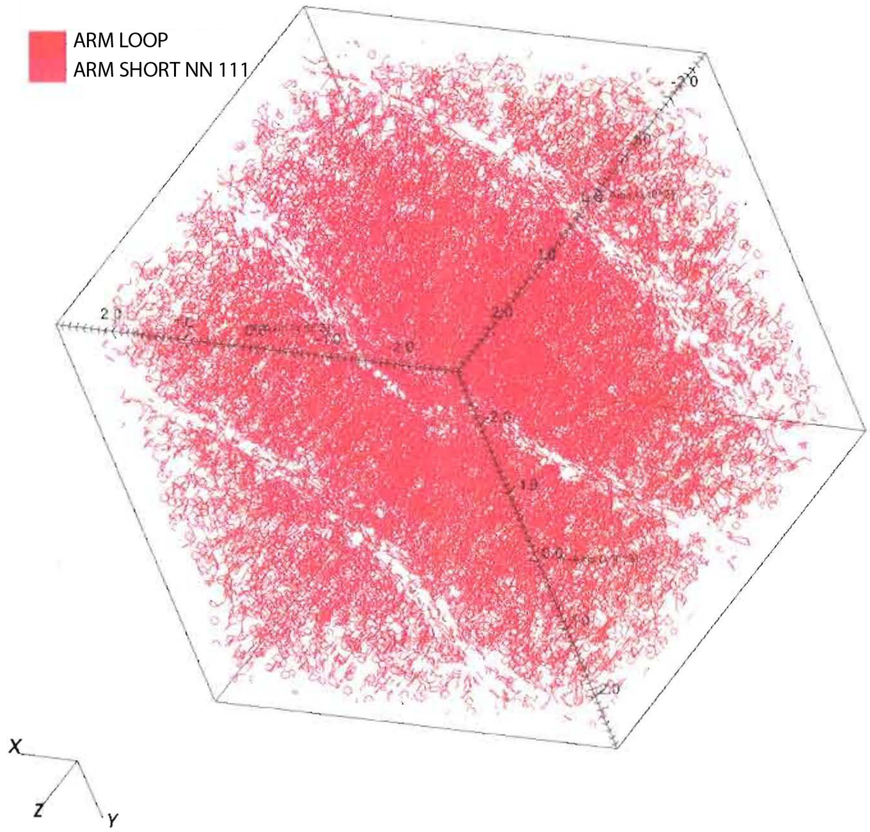

Figure 8. Results from the parameter-free modeling of crystal plasticity under extreme deformation.

That multiple models for material degradation exist and are under development should allow model cross-validation and iterative reduction in uncertainties and can lead to enhanced trust in the reliability of accelerated irradiation tests as a predictor of in-reactor material performance. The iterative approach proposed in this initiative focuses the attention of TSM developers on improving the accuracy of existing expedient models and on improving the computational efficiency of more accurate simulations. UQ provides a formal framework guiding and prioritizing ongoing developments in TSM so that uncertainties are quantified, propagated, and managed across all the material scales, from largest to smallest, relevant for property degradation.

4. Ion-Beam Capabilities and Limitations

Gary S. Was, University of Michigan

Ion irradiation has the potential to significantly impact the development of materials for both advanced light water reactors and Gen IV reactor concepts. A number of important irradiation effects were either discovered by or have been understood through the use of ion-irradiation experiments. These include the development of the theory of radiation-induced segregation, alloy stability under irradiation, the importance of the primary recoil spectrum to freely migrating defect production, the understanding of the void lattice, and the role of localized deformation in the irradiation-assisted stress corrosion cracking (IASCC) process.

The attractiveness of ion beams as a tool to understand radiation effects has always been the capability to conduct ion irradiations in a short amount of time, to high dose and thus at a very low cost. Depending on the ion and the nature of the ion source, irradiations can be conducted at displacement-per-atom (dpa) rates as high as 100 dpa/day, allowing experiments to probe the high-dose regimes of reactor core materials in a very short time. Electrons and heavy ions produce no radioactivity, and light ions result in only low levels of residual radioactivity. Thus, in most cases, samples can be handled as if they were unirradiated, greatly reducing the time and expense for post-irradiation analysis. Both of these features add up to greatly reduced experimental costs. In addition, ion experiments can be extremely well controlled in terms of dose, dose rate, and temperature.

Yet, ion irradiation and neutron irradiation differ in significant ways, making it incorrect to assume that ion irradiation is a direct substitute for neutron irradiation. While the neutron energy spectrum is very complicated and depends on the specific reactor type, ion beams are essentially monoenergetic sources and will produce different initial recoil spectra. Charged particles also have much greater cross sections for interaction with atoms than do neutrons, resulting in much higher damage rates, and therefore much shorter penetrations distances, per incident particle. As a result, ion beams give up their energy in very shallow layers compared to neutrons.

The type of ion has a large impact on the nature of the resulting damage (Figure 9). Electrons produce only isolated Frenkel pairs, while heavy ions produce dense cascades, and light ions produce smaller, more widely spaced cascades. The resulting point defect concentrations and damage morphology can, therefore, differ significantly among the various ion types. In addition, ion irradiation does not result in transmutation that produces elements such as He, a process that can be important to the development of the microstructure.

Nevertheless, ion irradiation has been quite successful in reproducing many of the critical features of the neutron-irradiated microstructure. In one set of experiments on a *common* heat of commercial purity, 316 stainless steel, both the magnitude of the "W"-shaped radiation induced segregation (RIS) profile for Cr as well as its spatial extent was shown to be nearly identical whether irradiation was conducted in a BWR core or with 3-MeV protons to the same dose. (Was, 2002) The agreement extended to radiation-induced hardening, phase formation, void formation, the dislocation loop microstructure, and the susceptibility to IASCC. Both proton and electron irradiation of model reactor pressure vessel steels produced hardening similar to that from neutron irradiation of the same alloy to the same dose. And heavy ion irradiation of a UMo-Al fuel clad couple revealed the formation of a single intermetallic compound, in agreement with neutron-irradiation results on the same sample geometry. (Palancher et al. 2009)

Ion irradiation also holds the promise to extend experiments beyond simple, single-beam irradiations. In both fission and fusion systems, transmutation is important in understanding the material response to irradiation. The use of heavy ion irradiation to produce the damage, simultaneously with injection of H or He, has been shown to be an effective means of accounting for H or He production in reactors. The use of energy degraders permits the tailoring of irradiation conditions to reproduce mechanisms that occur in reactor core materials. Furthermore, ion beams provide the capability to address the simultaneity of multiple components in the extreme reactor environment. While much emphasis is placed on the effect of irradiation, the combination of two or more of the key elements of the reactor environment—radiation, temperature, stress, corrosive medium—is critical in understanding degradation in these systems. Ion beams lend themselves very well to experiments such as irradiation-induced creep (high temperature, stress, radiation), irradiation-accelerated corrosion (high temperature, radiation, corrosive medium), or real-time tracking of radiation damage morphology in a transmission microscope (high temperature, radiation). Experiments using ion beams can be conducted in real time using diagnostics to track degradation as it occurs.

Ion beams offer the potential to provide greatly accelerated irradiation to study the performance of materials under complex environments at greatly reduced cost. Additional work is required to more fully understand the damage state resulting from different types of particles at different damage rates, so that the predictive capability of ion irradiation can be maximized for extrapolation to the high doses and extreme conditions of both advanced LWR and Gen IV designs.

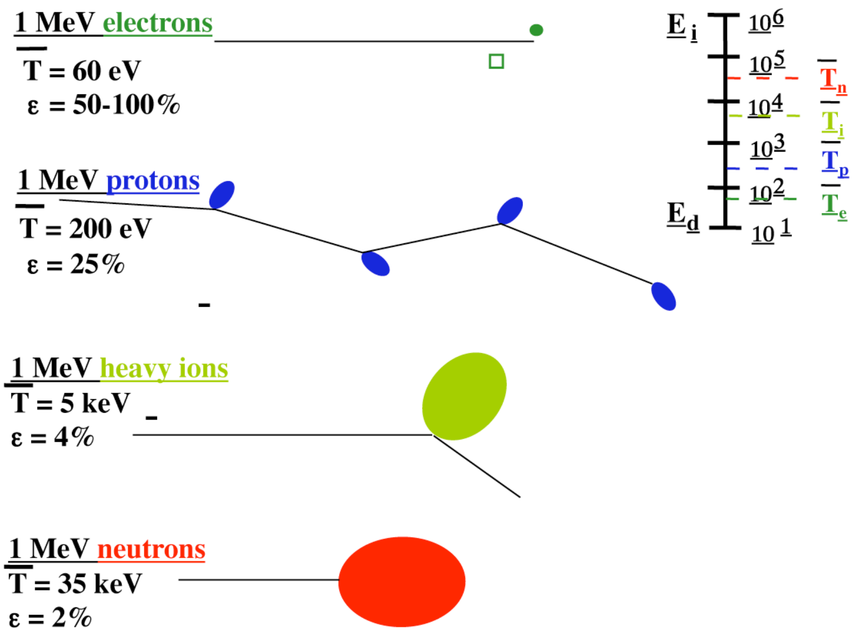

Figure 9 Differences in recoil energy and cascade morphology among various particle types. (Was 2007)

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5. Topical Area 1: Model Development

Tom Arsenlis, Lawrence Livermore National Laboratory

Overview

Develop a validated multiscale model that predicts the yield point and hardening rate of a neutron irradiated alloy using UQ to integrate theory, simulation and modeling with accelerated experimentation

Problem Statement

The long lead times for the deployment of new materials for critical components in nuclear reactor cores is directly related to the exclusive reliance on reactor irradiations for materials evaluation, and a lack of confidence that materials modeling and simulation and out of core materials testing can be used to quantifiably predict the performance of materials in those environments. The purpose of this topical area is to evaluate and demonstrate the maturity of multiscale materials modeling and simulation, and multiscale accelerated out-of-core experimentation for nuclear materials in developing a robust constitutive model for the irradiation strength and strain hardening under a variety of reactor conditions. A maturity test is proposed of the methodology focusing on a material for which there is high quality neutron-irradiation performance data that is kept unknown to the team developing the constitutive model with access to the unirradiated virgin material, ion beam and other out-of-core experimentation and characterization facilities, state of the art multiscale materials modeling tools and computer resources, and robust uncertainty quantification tools for the forward quantification of model uncertainty and parameter sensitivity.

Need

There is a need to understand the extent to which the life of the current LWR reactor fleet can be extended. These life extensions rest primarily on the property degradation of irreplaceable materials subject to prolonged irradiation. Current life extension program rely on materials monitoring and credible predictions of the end of life are difficult with current methodologies. Waiting for an old reactor to fail, and projecting lessons learned from that failure to other operating reactors is an unacceptable path forward.

There is a need to understand the behavior of materials in new reactor designs whose core conditions will significantly differ from previous designs in pursuing license applications for these designs. This is a classic chicken or the egg problem in which materials are necessary to build the reactor and the reactor is necessary to test materials. The result has been that the performance of old reactors is still being increased by uprates because of the overcautious operation in the past. This focus area has the potential to break this cycle and allow reactors to reach their operating potentials sooner.

Challenges

It is a challenge to identify a candidate material that has been irradiated to high doses that has adequate documentation for its initial processing, its thermal, mechanical, and neutron-irradiation history. Also, there must still be virgin material that has never been irradiated available. This study may return the result that although materials modeling and simulation has advanced significantly in the past couple of decades is still not mature enough to translate information across the four decades of time scale from ion to neutron irradiation. The causes of this may be that the time scales are to disparate to transcend algorithmically or that the sensitivities of model parameters to these different regimes of irradiation are so disparate that the it is not possible to project with low uncertainty information obtained with accelerator driven ions into neutron environments. However, even this null result would guide the strategic direction of future reactor materials research investments.

Impact

This topical area will demonstrate whether the materials modeling and ion-irradiation communities have matured to the extent where their tools and expertise have risen to the level in which their products can be used in practical nuclear engineering component design. All of the other topical areas within this report predicate their success on the success of this topical area. Success will lead to a reduction in the long lead times for material evaluation, qualification and deployment in nuclear reactors. Success will lead to a path forward in materials selection and qualification for new reactor designs before they have been built.

6. Topical Area 2: Chemistry and Property Evolution of Fuels

Patrice E. A. Turchi, Lawrence Livermore National Laboratory

Overview

Develop a multi-physics model to predict site-redistribution and fission-gas release, and their consequences on thermal conductivity of metallic U-Zr and U-Pu-Zr fuels irradiated to 20 at. % burn-up with operating temperature in the central region of the fuel pin up to 750°C using UQ to integrate theory, simulation and modeling with accelerated experimentation.

Problem Statement

The research objectives identified in the DOE-NE roadmap require the development of safe and reliable advanced fuel forms with higher burn-up (U.S. DOE-NE 2010). This objective is an integral part of the Fuel Cycle Research and Development (FCRD) fuel campaign with the development of advanced fuels using a goal-oriented science-based approach (U.S. DOE 2009). This approach relies on effective integration of small-scale, phenomenological testing with theory, simulation, and modeling to partially replace the classical heavily empirical (prototype-based) fuel qualification process that is lengthy, expensive, and that does not always provide optimum performance (Crawford 2007). In this context, ion-beam irradiation experiments combined with TSM and guided by uncertainty quantification (UQ) analysis for linkage to neutron irradiation and to fission effects, as proposed in this initiative, is one of the tools that may enable the science-based development. Once the models reach a level of prediction confirmed by validation and verification, the use of UQ may be a useful tool to integrate the overall fuel development activities up to the technical level of readiness where fuel qualification can be initiated.

Need

There is a need to assess fuel performance by accelerated probing of unit mechanisms operating in fuels under reactor conditions to deliver in a timely fashion critical information on fuel properties, notably in the areas of (1) thermodynamic properties under irradiation condition (driven systems far from equilibrium condition with, e.g., second phase precipitation), kinetics of formation and transformation under irradiation condition (e.g., enhanced diffusion), and (3) microstructural changes under irradiation condition (e.g., changes in grain size and morphology). In all three cases, ion-beam irradiation are ideally suited for identifying structural changes, quantifying enhanced diffusion and microstructural evolution, and hence, guiding the development of appropriate science-based models, monitored by UQ analysis, that need to be incorporated in fuel performance codes. Once the controlling factors that are key to fuel performance, based on UQ analysis, are identified with accelerated probing, the resulting science-based models can be exercised and validated against archived and new data from in-pile test-reactor experiments.

This initiative proposes to select a well-characterized neutron irradiated metallic fuel (e.g., U-Zr, U-Pu-Zr) as a base-line for which well-documented archived data and un-irradiated samples of the same batch are available, so that ion-beam irradiations can be designed and performed on comparable samples, and post-irradiation examination can be executed for addressing the 3 selected properties (see above) and their impact on mechanical integrity and thermal transport. With this information models that explain and scale the in-pile and out-of-pile data, guided by UQ analysis, can be integrated in performance codes.

Challenges

- The models or "key unit mechanisms" needed to characterize the phenomena associated with stability, enhanced diffusion, and microstructural changes in systems driven far from equilibrium are necessarily complex, involving multiple and often coupled variables.
- The operative key mechanisms must be identified, modeled, and managed through science-based fuel design.

Impact

The success of this potentially new paradigm from experimentally driven to science-based approach, combined with accelerated probing and UQ for advanced fuel development would reduce timeframe currently necessary to design and test new and advanced fuel forms. This approach is also expected to provide a more adequate assessment on life extension and materials evaluation for advanced fuel concepts, and a better fuel management with more appropriate margins of error and better uncertainty quantification.

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7. Topical Area 3: Advanced Clad Materials Development

Michael Fluss, Lawrence Livermore National Laboratory

Overview

Utilizing intrinsically radiation tolerant materials does not in itself fully deal with the complexity of cladding materials for advanced high burn-up fuels nor does it address the issues associated with existing fuels in BWR and PWR reactors. The design and development of advanced clads may be the most challenging and the most important single issue of advanced reactor development since the clad is the principle containment vessel for the evolving fuel system and ultimately determines the service life of the fuel itself, the economics of the fuel cycle, and the nature of the consequential disposition strategy.

While radiation tolerance is a key element in the technology challenge for advanced claddings, it is important to design the materials to withstand the challenges of temperature, corrosion, stress, resulting either from external surface stressors, such as hydrogen production from hydrolysis in BWRs, or from internal stressors such as fission gas pressure and expanding fuel volume, corrosion from fission products such as Pd, as well as phase transformations and deformation associated with the action of the various thermal, mechanical, and radiation driving forces on the clad bulk material itself.

Problem Statement

The goal of high burn-up fuels raises a significant challenge to the development of advanced claddings. The challenge is that to study fuel assemblies will require many years in the relevant reactor environment, followed by cooling, and subsequent materials characterization analysis. New ideas for fuel cladding must ultimately be studied in the presence of the various stressors and under conditions of radiation that can focus on the unit mechanisms responsible for key degradation processes.

Hydrogen pickup significantly reduces cladding ductility for normal operation and accidents, Irradiation significantly reduces the macro-ductility of zirconium alloys to nearly zero while the micro-ductility remains high . The prediction of fuel failures due to cladding deformation has large uncertainties because the mechanisms for micro-deformation are not understood

Need

- Understand and model hydrogen pickup for waterside corrosion on a micro-scale for application to macro-scale.
- Understand and model micro-deformation for predicting failures on a macro-scale.
- The fuel-clad interface is arguably the place where the rubber hits the road and is among the most complex challenges of materials predictability under reactor

conditions. To understand this interface one must understand the combined effects of the complex evolution of the fuel (the production and migration of corrosive fission product elements, and the internal pressurization of the cladding resulting from swelling of the fuel and the associated production of fission gasses) along with the evolving properties of the clad itself (resulting from displacement damage, hydrogen uptake, and the production of transmutation products such as helium).

Challenges

- Model and understand why some cladding types pickup different levels of hydrogen due to waterside corrosion
- Model micro-deformation for application on a macroscale
- Understanding "bulk" behavior (e.g., creep, growth) using ion irradiation which has limited penetration
- The models needed to characterize the phenomena are necessarily complex but can be broken down into studies relevant to specific diffusion couples and phase stability issues in the presence of transmutants and fission products.
- Since the principle issues are associated with a combination of thermodynamics and kinetics, techniques such as CAPHAD, DICTRA, and phase-field methods would be employed to the advantage of this topical area.

Impact

- This approach will lead to identifying and isolating the key unit mechanisms.
- Being able to model and understand the micro-deformation and hydrogen pickup will assist in developing alloys that can have higher ductility in addition to a more accurate prediction of failure
- Could lead to a step-change in our understanding of the most important degradation mechanisms
- Accelerated investigation of the cladding-fuel interface, and the cladding coolant interface under conditions of irradiation, thermal, and mechanical stress will lead to a more rapid development of advanced cladding materials by allowing new ideas to be investigated and if found to be deficient, dismissed early in the development process. On the other hand, promising approaches can be studied in detail, varying relevant parameters.

Participant Reaction

The workshop participants identified that accelerated experimentation involving intrinsically radiation tolerant materials focused on the fuel-cladding interface driven by accelerated ion-beam studies could significantly accelerate the development of new ideas for advanced cladding materials.

Worthiness of the Challenge

Developing advanced cladding material is a foundation for NE's long-term fuel cycle strategy for improved nuclear energy system performance of safety, efficiency, and cost.

Participant Suggestions

Stuart Maloy (LANL) noted that the challenge was identifying and modeling unit effects leading to radiation damage in core materials and developing and validating models using archive data and specimens as well as new data from a range of irradiation sources. A successful program, presently being addressed in FCRD's core materials program under advanced fuels and some university research under the NEUP programs, would significantly reduce the timeframe to develop, test and qualify materials for high dose nuclear applications and result in the development of models that accurately simulate radiation effects in cladding materials to understand and predict these effects in engineering alloys.

It was noted that extending the effort to include accelerated studies of the fuel clad interface would lead to the compromises and optimization required of a truly useful technological fuel clad material.

Revised TA

Predict yield point and hardening rate, including irradiation creep effects of cladding irradiated to high dose using UQ to integrate theory, simulation and modeling with accelerated experimentation. Begin with zircaloy clad at temperatures in the range 290-360C and extend the methodology and apply it to clad for advanced reactors.

8. Topical Area 4: Fuel-Clad Interaction Leading to Fuel Failure

Gary S. Was, University of Michigan
Wayne King, Lawrence Livermore National Laboratory

Overview

The original task was to model high burn-up, high power density failure modes of zirconium alloy cladding in contact with oxide fuel under the following environmental conditions: operating temperatures 290–360°C and LOCA conditions to 1000°C, water chemistries including Li/B-containing PWR coolant and BWR normal and hydrogen water chemistries, temperature, and stress using UQ to integrate theory, simulation and modeling with accelerated experimentation.

Problem Statement

The lack of predictive modeling capability for fuel performance and reliability will limit the performance of LWRs, especially at higher burn-up and for reactors undergoing power uprates.

Need

- A model for the performance and failure of Zr-clad UO2 in BWRs and PWRs with the capability to handle power uprates and high burn-up.
- The capability to handle various degradation modes, such as SCC due to PCI, fretting, debris, etc.

Challenges

- Insufficient understanding of microstructure and microchemistry evolution of both fuel and clad and their interaction.
- Modeling a discrete event, such as failure, especially given that multiple failure modes are likely to exist.
- Extension to fast reactor and TRISO fuels.

Impact

- Quantify fuel lifetime predictions to maximize energy output of fuel
- Quantify margins and build the case for power upgrades and higher burn-up targets.
- Guide development of key inspection practices and provide insight to fuel modeling for advanced reactor designs.

Participant Reaction

Participants agreed that this TA was both a very challenging and worthy goal, especially if expanded to encompass the full range of relevant failure modes of LWR fuel.

Worthiness of the Challenge

The importance of being able to model fuel failure is generally regarded as an important goal as it affects the operation and economics of light water reactors, especially as utilities continue with power uprates and to push burn-ups higher.

Participant Suggestions

Jeremy Busby noted that fuel behavior/fuel failure is not a life extension issue. Michael Billone suggested that fuel-cladding bonding and the development of a sub-micron, Pu-rich fuel rim are processes that could be important in fuel failure. It was also noted that the deformation mode of irradiated zirconium alloys is characterized by dislocation channeling, which is different from the unirradiated condition.

Revised TA

Use a combination of UQ, TSM, and accelerated experiments to develop models for the performance and failure of zirconium alloy-clad, oxide fuels at high burn-up and high power density under operating and off-normal conditions and extend the methodology and apply it to cladding failure in advanced reactors.

9. Topical Area 5: Design a Radiation-Tolerant Material

Todd Allen, University of Wisconsin
Michael Fluss, Lawrence Livermore National Laboratory

Overview

Design and develop materials that mitigate or possibly eliminate the negative effects on performance of the extreme irradiation environments while maintaining fabricability. Uncertainty Quantification (UQ) will be used to integrate theory, simulation and modeling with accelerated experimentation. The resulting materials will exhibit smaller and more predictable variability in properties and performance during their in service life.

Problem Statement

The goal of closing the nuclear fuel cycle is dependent on achieving higher burn-up of the nuclear fuel so as to minimize fuel reprocessing. The cost and time necessary to develop and license new nuclear materials is prohibitive. New materials that are radiation tolerant and exhibit smaller uncertainties in in-service properties are needed for the successful development of advanced thermal and future fast reactor technology.

Need

- Modeling and experimentation techniques that enable mechanistic-based approaches to technology development will accelerate materials development
- A successful science based approach to the development of radiation tolerant materials will lead to the deployment of engineering materials.

Challenges

- The models or "key mechanisms" needed to characterize the phenomena associated with radiation tolerance (helium management, dislocation dynamics, etc.) are necessarily complex, involving multiple and often coupled variables.
- The operative key mechanisms must be identified, and managed through science based materials design and engineering.

Impact

- Accelerate innovation and down-selection of design and material options, thereby reducing development times to bring new materials to market
- Identify and design new materials options for future reactors

Participant Reaction

A science-based approach that can accelerate the development of a radiation-resistant material is a good test case for the UQ, TSM, and accelerated experiment methodology. The TA example of high temperature He embrittlement in austenitic alloys is a good test case, although other material classes or specific degradation mechanisms could also be chosen. Cost and fabricability must be carefully considered if the resulting radiation tolerant material is to be technologically relevant for nuclear energy applications.

Worthiness of the Challenge

Developing a radiation tolerant material is a foundation for improved nuclear energy system performance (safety, efficiency, and cost).

Participant Suggestions

Rick Kurtz from PNNL added two suggestions. First, he noted that the fusion programs have a set of models for understanding He-embrittlement that could be used as a starting point for the UQ analysis. He also noted that they may have samples that have already been irradiated that could be used as a starting point for the topical area.

It was also suggested that the UQ methods be applied in making an intelligent down select of the materials type, austenitic, ferritic or ferritic-martensitic, or oxide dispersion strengthened (ODS), taking into consideration fabrication issues and materials costs and fuel cladding interface failure issues.

Revised TA

The use of a combination of UQ, TSM, and accelerated experiments to design an austenitic material that has significantly increased resistance to high-temperature He embrittlement is suggested. Achieving this goal would allow improved material lifetimes for metallic components in light water reactors, high temperature gas-cooled reactors, and liquid metal cooled fast reactors while maintaining fabricability. Uncertainty Quantification will be used to integrate theory, simulation and modeling with accelerated experimentation. The resulting materials will exhibit smaller and more predictable variability in properties and performance during their in service life.

10. Stakeholder Discussion

The stakeholder discussion session focused on identifying the initiative's potential benefits to various organizations. Speakers from the nuclear power industry and relevant government agencies provided a brief overview of current operations and described future long-term needs in the area of nuclear energy materials. They also identified research areas that were most pertinent to the work and explained the importance of breakthrough discoveries might have to their work.

This section summarizes the presentations given by the six speakers. Representatives from two organizations, the Electric Power Research Institute (EPRI) and GE Global Nuclear Fuels, could not attend the workshop but provided letters, which are reprinted at the end of this section.

General Atomics

Bob Schleicher, a reactor designer in the Fission Group at General Atomics (GA) noted that his group and the Magnetic Fusion Group, represented at the workshop by GA Vice President Ron Stambaugh, are interested in research that will push the boundaries on radiation-resistant materials. The EM^2 gas-cooled fast reactor concept under consideration by the Fission Group will require structural and clad materials that can survive fast neutron irradiation for up to 30 years. The group is considering a silicon carbide composite that, if success, could significantly increase uranium utilization and help address problems associated with reactor waste. Combining an accelerated aging technique such as triple-ion beam acceleration with an advanced modeling capability such as UQ would allow researchers to better understand the composite material's behavior over time and could decrease the time needed to develop such breakthrough materials.

The Magnetic Fusion Group is also working to develop radiation-tolerant materials and, in particular, needs a first wall material that can survive more than five years. The proposed initiative, by advancing work to develop such a material, could help make magnetic fusion a reality.

Westinghouse Electric Company, LLC

Randy Lott discussed the research areas of interest at the Westinghouse Materials Center of Excellence. Westinghouse is now considering operating its power plants for 60 years and beyond—20 years longer than originally planned. In the process, materials scientists must continue to "bravely go forward" into new operating territory by extrapolating performance based on past data and what researchers know about material behavior. Lott noted that progress in reactor technology has always been accelerated (Figure 10). The neutron was theorized in 1920 and discovered in the 1930s. Scientists achieved the first chain reaction in 1945. Archival data on 60-year-old irradiated materials is not available; scientists would have had to start such experiments in 1950 to have the experimental data they need today. Licenses for the first commercial power plants were issued in 1970s.

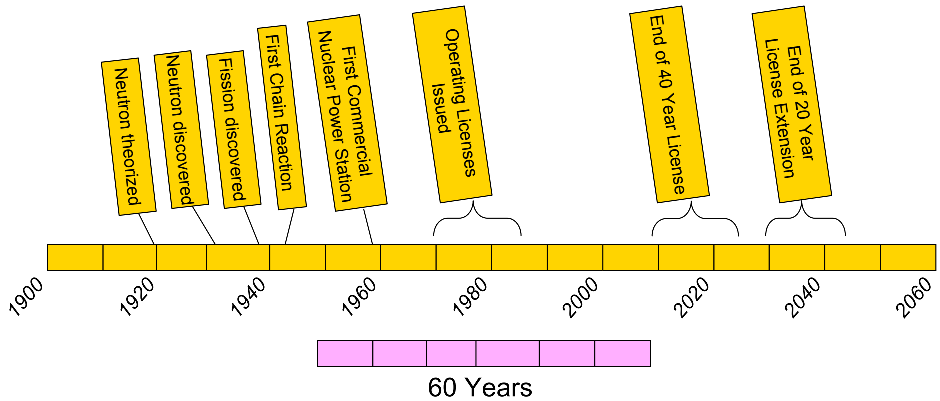

Figure 10. Timeline showing accelerated progress in reactor technology.

Westinghouse wants to extend those 40-year license agreements. As a result, the company is extremely interested in better understanding the issues involved in long-term irradiation of various materials.

The proposed initiative is combining simulation, modeling, and field experience to develop a three-pronged approach to help advance materials research for nuclear energy applications. On the commercial side, researchers tend to, instead, use information on materials, environment, and structure to determine how long a component will last and how well it will perform. The tools must be used to examine the structural and environmental elements of component lifetime, not just material parameters, because all three factors have built-in uncertainty.

Lott also noted that most of the data on irradiation environments is from fast breeder/fusion studies, where materials are exposed to hundreds of displacements per atom and high fluence situations. The known issues under these conditions are material swelling, loss of ductility, loss of fracture toughness, and creep—all problems that must be managed to ensure long-term performance. Gen IV reactors have similar problems because they, too, are operating at high temperatures.

Light-water reactors operate in a slightly different regime, in particular, at lower temperatures and within a different dpa/neutron spectrum. Some of the same problems occur in LWR materials, for example, loss of ductility and fracture toughness, stress relaxation, irradiation stress corrosion cracking. However, the operating environment in LWRs ranges from 0 to 100 dpa, with many components experiencing only 10 dpa. Thus, Westinghouse researchers need to analyze material at these in-between doses but with irradiation occurring over a 60-year timeframe so they can understand how the rate effects play into material performance. For example, scientists can easily simulate the environment of an LWR pressure vessel operating at 10^{-3} dpa/s. What's important is determine how they can extrapolate forward in time and know that they are not moving materials into an entirely new regime. Therefore, Lott emphasized that an important part of the proposed initiative must be developing an understanding of how simulation, modeling, and experiments come together to influence analyses.

The topical area on nuclear fuels has the potential for more immediate contributions to reactor research at Westinghouse. Nuclear fuels are an active production line, and improved materials can be used in existing reactors now, provided they are compatible with those facilities. Having advanced modeling capabilities does not, itself, improve fuel performance. Models merely describe that performance. However, uncertainty in material performance almost always limits a plant's operations. Reducing that uncertainty can increase the range of acceptable operating conditions. Models may also indicate a new approach to improve operations: For example, a fuel might perform better if it is loaded differently or it receives high fluence at the first or last step in the process. Improved models are also valuable if they allow researchers to improve a fuel's design. The problem is complex and involves many interactive models, issues that Lott notes are addressed in the discussion of Topical Area 4.

Another area of concern for Westinghouse is its reactor pressure vessels where there is an abundance of surveillance data. Many uncertainty quantification and uncertainty propagation issues are related to continued operation of this aging fleet. For example, uncertainty in the composition, radiation environment, radiation temperature, or radiation history can affect predictions of future performance. More important is to develop methods to address signals in the data that indicate a future problem, such as embrittlement mechanisms that have not been modeled beyond 40 years. Lott noted that low copper forgings and other advanced materials have improved the designs for new fleets such that reactor pressure vessels are not likely to be a problem in the future.

One challenge in the modeling area is that most models are empirically based or mechanistic. Yet, modeling only one mechanism does not answer all the questions because material behavior is affected by two, three, or four mechanisms operating simultaneously. However, the regulation system for the nuclear power industry is based on probabilistic analysis or uncertainty propagation. These codes, which address probabilistic risk assessment and probabilistic fracture mechanics, may not have the mathematical structure of the UQ models discussed at the workshop, but they may be valuable to the proposed initiative.

Developing new materials for reactor components are not only an issue for new reactors but also for replacement materials in the aging fleet. Damaged components must be replaced, and Westinghouse researchers often need to develop new alloys for replacement components. Models that predict irradiation stress corrosion, embrittlement, and creep can thus help maintain the existing fleet. The challenges in developing these models are that data come field components—not archived experimental research. In addition, extrapolations must be made using the fast reactor data and the limited (and expensive-to-generate) irradiated material.

Lott concluded by noting that in advanced technology efforts such as the proposed initiative, it is incredibly important to set goals high enough so that researchers are challenged to think about something truly different.

DOE Office of Nuclear Energy

Sue Lesica, the workshop sponsor, noted that NE is looking for revolutionary changes in the area of nuclear energy materials. Small, incremental changes will not provide the materials needed to meet the nation's future energy demands. In addition to these high goals and high expectations, NE also has the luxury of time to develop revolutionary materials, as opposed to commercial vendors such as Westinghouse. In addition, because NE does not have a fast reactor or FFTF for testing new materials, NE is interested in using any available tool to push the state of the art in materials science.

Fusion Energy Sciences, DOE Office of Science

Materials are a significant challenge for Fusion Energy Sciences (FES). Gene Nardella, who is responsible for materials research at FES, noted that the program's new management recognizes the need to move forward faster in this area because it is such a significant challenge. In that regard, FES wants to establish better partnerships with NE, because of the similar problems, as well as with NNSA and BES. One if the biggest concerns is obtaining experimental information. The program has effectively used current tools such as fission reactors, but it does not have a fusion-relevant neutron source. The program is exploring mechanisms for obtaining such a source, but Nardella notes that it is not cheap. As a result, FES is very interested in learning about any tools—whether from the proposed initiative or other programs—that will help researchers better understand and develop the materials needed to withstand a fusion environment.

Naval Reactors

Naval Reactors (NA-30) is an engineering-oriented operation with cradle-to-grave responsibility for the Navy's nuclear reactors. Today, the Navy has the same number of operating reactors as the U.S. commercial nuclear industry. Naval Reactors is responsible for research and development on the next generation of reactors, design and oversight of the construction of new reactors, maintaining and operating reactors in the fleet, and disposal of spent reactor cores.

John Hack (Bettis Atomic Power Laboratory) noted that Naval Reactors has an interest in tools that would help do these jobs better, particularly results-oriented work that leads to better models and better predictive tools. Naval Reactors' primary concerns are LWRs and developing advanced reactor materials. Hack discussed potential synergies with the proposed initiative, based on a draft white paper received before the meeting, which described the high-level objectives of the initiative but did not discuss the expected scope, detailed work tasks, or detailed program plans. Figures 11 and 12 show the slides presented by Hack at the workshop and are followed by a brief summary of his talk.

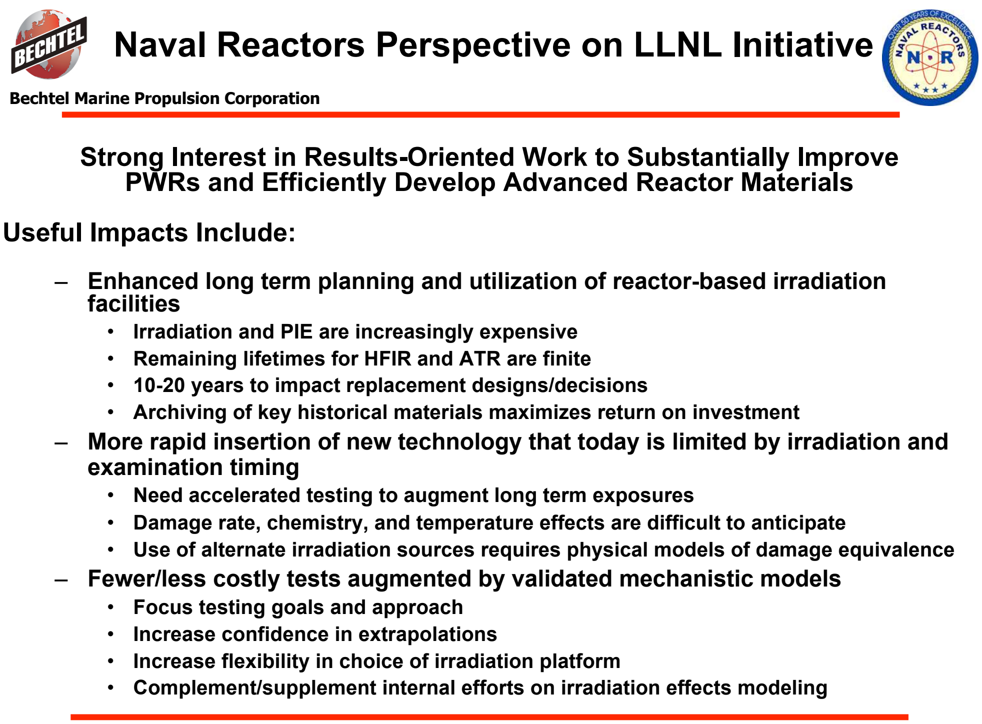

Figure 11. Slide 1 from the stakeholder presentation for Naval Reactors.

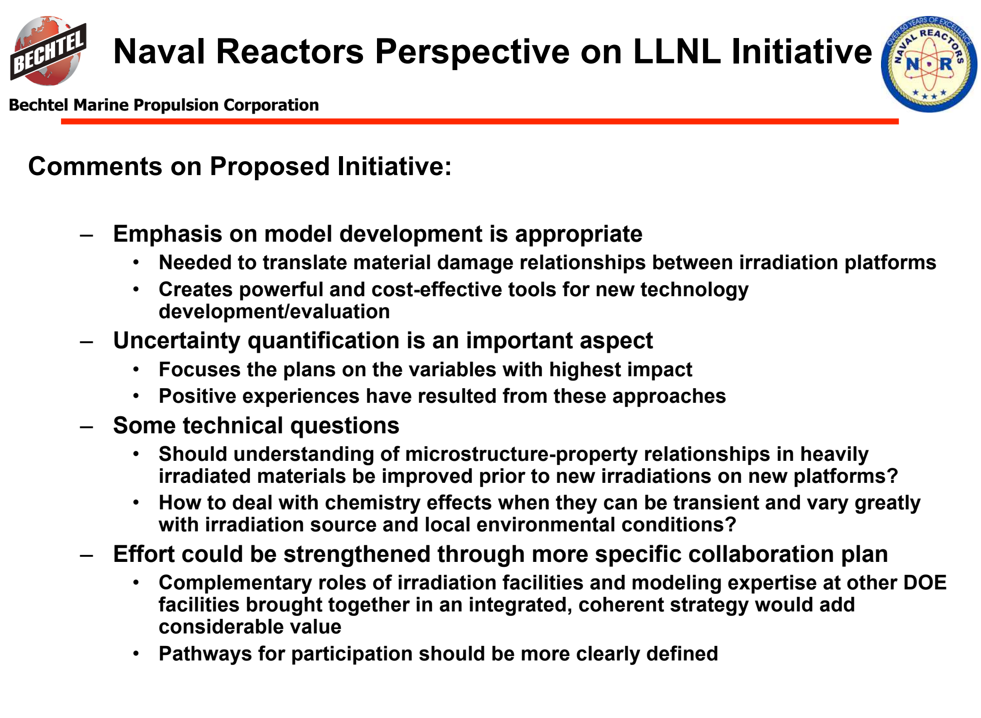

Figure 12. Slide 2 from the stakeholder presentation for Naval Reactors.

In general, the Naval Reactors Program has relied on in-reactor experiments to predict reactor material and component performance. Better predictive tools could reduce the reliance on testing and lead to different, less-expensive types of tests. Such tools could also be useful in defining the functional requirements of the next-generation US test reactors, which need to be established within the next 20 years.

Unlike commercial reactors, some naval reactor cores must remain in service throughout the lifetime of the reactor plant. This requirement imposes the need for accurate predictive capabilities and erects a high bar for inserting new materials technologies. Efforts proposed in the initiative—such as accelerating testing through the use of ion-beam irradiations, improved physical models for the evolution of irradiation damage and its impact on material performance, and uncertainty quantification for guiding research efforts and providing a framework for risk management—are all of interest, if successful.

Regarding the proposed initiative, the emphasis on model development appears appropriate. Having the ability to translate materials damage relationships between test platforms would provide some powerful tools, but others have tried before with limited success. In designing the ion-irradiation experiments, the initiative proponents must understand the quality of the data to be compared with results. For example, data are available on zirconium alloys at various fluences, but the wide variation in material and exposure conditions means the data are not necessarily self-consistent. Also, there is little agreement within the community on specific deformation mechanisms, let alone a unified model for radiation creep or in-pile deformation that takes into account all the mechanisms at one time. Chemistry effects are another concern because behaviors tend to be transient and dependent on irradiation type.

Finally, Hack noted that integrating this work with the various irradiation facilities and expertise across DOE sites would allow for more efficient progress; however, the draft white paper did not clearly define the opportunities for collaboration or for participation by external groups.

Q: I had a question about the next generation of naval reactors. Do you see your future needs as being different from energy facilities? When people talk about Gen IV or breeder reactors, [the work they're talking about?] is pretty aggressive. I'm guessing that in your world, the next-generation reactor is likely to be evolutionary rather than revolutionary. Do you even think about that?

A: We definitely think about it, but again, the bar is very high. We have 60 years' experience running what we have. For current needs and for projected needs, we're in pretty good shape with what we have. However, we're always trying to push out. We've had programs come and go, both revolutionary and evolutionary. We've tended toward the evolutionary when we actually apply things, but we've certainly looked at revolutionary concepts and continue to.

Nuclear Regulatory Commission Office of Research

Robert Tregoning, a technical advisor for materials in the Division of Engineering at the Nuclear Regulatory Commission Office of Research, discussed how the needs of NRC align with the proposed initiative. He noted that NRC has some of the same interests as GE, EPRI, and Westinghouse, especially in terms of long-term operations, the performance of reactor internals, and pressure vessel materials as we move into longer times and longer operation periods than we have experience or surveillance information to determine if new mechanisms or phases occur.

In addition to the reactor materials mentioned in earlier presentations, NRC is interested in other materials that experience radiation at extremely low dose rates but for long time periods. Such materials include containment coatings and other coatings used in the commercial plants, the containment materials themselves, concrete, rebar, metal structures, and cables, whether they are metallic, nonmetallic, and polymeric. Because these components suffer radiation damage at such low levels, the synergistic effects may be just as important or more important than a single mechanism. Researchers must therefore consider thermal and other effects of environment and how those effects interrelate with the irradiation effects.

NRC has a long-standing, robust research program in fuels and is interested in pushing burn-up factors so that more effective and efficient fuels can be developed. Another area of interest is how fuels perform under both normal and severe accident conditions. The proposed initiative may also shed light on various regulatory issues. NRC wants to move from what is now a prescriptive-based regulatory system to one that is more performance-based. Performance-based regulations would address a more diverse classification of fuels than the current zirconium-based fuels.

Gen IV materials are also of interest. Tregoning noted that the spectrum for ion-irradiation and other fast reactor programs is very consistent. In addition to high-temperature metallic materials, NRC wants to evaluate the performance of graphite components because graphite is used as a moderator and a structural material in many Gen IV concepts. The proposed work on fuels and cladding for advanced reactors would be valuable for NRC as well.

NRC is interested in UQ but has not applied it to materials. NRC researchers are developing probabilistic models, for example, to evaluate structural integrity and component performance over long lifetimes. They also want to do rigorous quantification of uncertainty to understand how accurate their models are. Such models help scientists not only to understand the risk associated with component performance, which is valuable in an absolute sense, but also to quantify and determine research priorities given the limited resources available. For example, if the failure risk for a certain component is largely consumed by some portion of a model, does that indicate a true risk or is it a reflection of the uncertainty in that portion of the model. When the problem is a reflection of uncertainty, resources can be directed toward research to quantify that risk. In fact, NRC is developing and using UQ-type tools to predict the structural integrity of primary components such as the system pipeline and pressure vessel and is moving into the area of steam generator II predictions.

ELECTRIC POWER RESEARCH INSTITUTE

April 28, 2010

Dr. Wayne E. King
Physical and Life Sciences Directorate
Lawrence Livermore National Laboratory
P.O. Box 808; L-353
Livermore, CA 94550

Subject: EPRI Interest in Ion Beam Capabilities

Dear Dr. King:

Thank you for the invitation to participate in the upcoming workshop between various industry stakeholders interested in ion beam applications and the Office of Nuclear Energy (NE). Although I will not be able to attend the meeting, I wanted you to know that EPRI strongly supports development of the use of ion beam irradiation coupled with theory, simulation, and modeling and integrated using uncertainty quantification to develop predictive models for material performance under irradiation.

Our interests at EPRI are generally aligned with classical applications of ion beams, i.e., to complement neutron irradiation studies to better understand the underlying mechanisms that impact commercial fuel cladding properties (strength, ductility, irradiation creep and growth, corrosion resistance, etc.). This interest corresponds with your campaigns 1 (time and length scale transcending models) and 3 (cladding).

Irradiation growth is a key performance characteristic in commercial LWR fuel and uncertainties in this area limit our ability to design robust, high burnup fuel. The interaction between hydrogen (from corrosion) and irradiation growth has been most recently connected with bowing BWR fuel channels which impacts control blade movement. Thus we are particularly interested in your campaigns 2 (cladding) and 3 (fuel/clad interface), including the uncertainty estimates that would come from the uncertainty quantification. We had quite interesting discussions with several LLNL experts earlier this year about a novel application of ion beam techniques to study the synergies of radiation damage and hydrogen in irradiation growth of zirconium alloys. In particular, an ion beam capability that could simultaneously irradiate a target material with heavy ions and a proton beam could independently vary two key factors in irradiation growth, namely point defects and hydrogen.

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Dr. Wayne *E. King*
April 28*, 2010*
Page 2

We would be very much interested in keeping up with the progress of this initiative and look forward to continuing this dialogue on commercial fuel applications.

Sincerely,

Kurt Edsinger, Ph.D.
Sr. Program Manager, Nuclear Fuel

c: Dr. Albert Machiels (EPRI)

Global Nuclear Fuel
A Joint Venture of GE, Toshiba, & Hitachi

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Dr. Wayne King
Lawrence Livermore National Laboratory
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Livermore, CA 94551-0808
May 10, 2010

Dear Wayne,

Thank you for contacting me regarding GNF participation in this week's Workshop on Accelerated Nuclear Energy Materials Development. Unfortunately, unforeseen work here in Wilmington prevents us from sending someone to participate in the Workshop.

We encourage and support in principle an initiative proposed "to use uncertainty quantification to integrate theory, simulation, and modeling with accelerated experimentation to predict the behavior of materials and fuels in an irradiation environment." Similarly, we encourage and support in principle a campaign approach for developing predictive models for fuels and cladding and new radiation tolerant materials. The time and cost associated with developing and licensing new materials is a considerable barrier to introducing new nuclear fuel technologies. Establishing techniques to more efficiently identify new materials and then to collect the performance data needed for licensing new technologies seems appropriate for DOE funding, and a task for which the Department's research community is well suited.

We believe the benefits of these efforts will not be realized for some time, but agree that addressing known barriers to enable tomorrow's technology is a worthwhile objective. In particular, we can envision benefits which reduce the time needed to explore and identify new candidate materials for commercialization. However, one challenge to bear in mind is the burden of proof required to shift regulatory processes and expectations away from licensing supported by performance demonstration at relevant exposure times.

Our resources for supporting these efforts are limited, but we look forward to participating when we can.

Sincerely,

DC Crawford

Douglas C. Crawford, Ph.D.
Manager, Fuel Performance & Design

Global Nuclear Fuel

11. Interactions, Interdependencies, and Synergies with Other Organizations

To demonstrate the efficacy of our approach, the initiative should initially focus on TA1 (methodology proof of principle) and TA5 (materials design and development) in the extreme of high irradiation dose. The greatest potential for interaction of the initiative with the existing NE program is in TAs 2,3, 4, and 5. It is suggested that, UQ experts and ion-irradiation capabilities from the initiative could be integrated with existing fuels projects (NEAMS) to test the utility of the methodology for fuels research.

The initiative is cross cutting and has synergy with industry (GA, Westinghouse, GE, and EPRI) and other federal offices including naval reactors, the Nuclear Regulatory Commission, and the Office of Fusion Energy Sciences (see stakeholder discussion). Industry, Naval Reactors, and NRC expressed support for this type of initiative and a general interest in collaboration in the area of light water reactor materials research. NRC indicated an interest in the application of UQ particularly for setting research priorities and moving to performance-based regulation. Basic research areas of importance for nuclear energy were identified through previous workshops (BES 2008; Roberto and Diaz de la Rubia 2006). There is also the possibility for the initiative to crosscut with the Office of Advanced Scientific Computing Research (ASCR), a possibility that was not discussed in the workshop but has been identified in a recent workshop report. (Adams et al. 2009)

References

Adams, Marv, Richard Klein, Paul Turinsky, Hany Abdel-Khalik, John Kelly, Jim Stewart, and Rizwan Uddin. 2009. Energy Systems Enabled by Advanced Modeling and Simulation at the Extreme Scale. In *Scientific Grand Challenges: Science Based Nuclear Energy Systems Enabled by Advanced Modeling and Simulation at the Extreme Scale*, edited by E. Moniz and R. Rosner. Crystal City, VA: Office of Science, Advanced Scientific Computer Research and the Office of Nuclear Energy.

BES. 2008. Basic Research Needs For Materials Under Extreme Environments.

Roberto, Jim, and Tomas Diaz de la Rubia. 2006. Basic Research Needs For Advanced Nuclear Energy Systems: Report of the Basic Energy Sciences Workshop on Basic Research Needs for Advanced Nuclear Energy Systems. Germantown, MD: Office of Basic Energy Sciences.

12. Workshop Recommendations

The workshop participants generally agreed with the approach of using UQ to integrate TSM with accelerated experiments. The questions that were posed regarding UQ were focuses on the quality of existing models, the poor quality of existing neutron data, and how UQ handles such issues as missing physics and unknown unknowns. These issues were addressed above in the section describing UQ. The participants saw the initiative as a high risk, high payoff activity. Participants also that identified interfaces with the other program elements (e.g., NEAMS, LWRS, FCRD Fuels and clad materials research, and research funded in NEUP) must be managed to ensure the efforts are coordinated and synergistic.

The three areas, UQ, TSM, and accelerated experimentation were seen as sufficiently mature to justify starting the initiative at this time.

Regarding the TAs, the question of whether the initiative should focus on LWR materials or on materials for advanced reactors was not resolved. After discussion with NE, the TAs were discussed and revised where appropriate. The revised TAs are included in the Topical Areas section above.

TA1 Recommendations

The workshop participants did not recommend any changes to TA1.

TA2 Recommendations

TA2 brought a range of comments from the participants. On one hand, it was recommended that we should not attempt TA2 because “limitations of understanding of the physics and the phenomena are so great that the unknown unknowns really dominate.” On the other hand, it was recommended that TA2 was a worthy endeavor because of the interactions between fuel and clad. In the end, it seemed that potential collaborations with the INL's fuel team increased the likelihood of successfully contributing to the development objectives, tipped the scale toward including TA2 in the plan. The question of whether to work on metal or oxide fuel was raised. From the viewpoint of TSM, we are in a much stronger technical position to tackle metal fuels at this time. It was also suggested that the costs may have been underestimated for TA2 considering what is currently spent in the NE program.

TA3 Recommendations

The question of whether the initiative will focus on LWR or advanced-reactor materials was raised. In addition, it was recommended that the LOCA aspect of the TA be dropped.

TA4 Recommendations

TA4 was seen as a challenging TA but one that is high pay off. One participant did indicate that this is not relevant for the current fleet. Fuel-clad interaction is highly relevant for the current fleet and for GEN3 and 3+ LWRs. The issues it addresses include increased burn-up, uprating of plants and reduced cost of fuel failures. It does not address plant life extension, as fuel is a "consumable." A participant said, "If you go back to FFTF design days, for a while, we tried to separate the fuel behavior from the cladding behavior. But they're two adjacent, interacting systems. Then you put the test into the reactor that has both fuel and cladding, and it responds differently to either one behaving independently. The whole interaction became known as the fuel adjacency effect. Everything you didn't understand became the fuel adjacency effect, which became for a while bigger than anything you did understand." It is difficult to separate out and ascribe processes to either the fuel or the clad as they're not only intimately linked, but their interaction gives rise to new phenomena that is not possible to discover by studying each separately.

TA5 Recommendations

TA5 had two specific recommendations. First, it was recommended that a specific application be selected and a goal set based for that application. Second, it was recommended that if the radiation damage models that will be used in the design of the material are not robust, that this TA should be delayed until models were developed in the other TAs. One participant placed this as the highest priority TA (see Appendix B: Questions and Answers)

13. Conclusions

Based on the questions that were asked at the workshop, the stakeholder discussion, and subsequent interactions with participants, the following conclusions have been drawn: The focus of this initiative should be on accelerating materials development through the synergistic employment of mechanistically focused high rate experiments coupled with advanced theory, simulation, and modeling leading to the design of new fission reactor materials for the extreme of high irradiation dose and high burn-up. Where neutron-irradiation experiments are not feasible, either because the appropriate spectrum is unavailable or because long irradiation times would be required, ion-beam experiments should be coupled with theory, simulation, and modeling and integrated using UQ to effect advanced irradiation effects scaling. The need for advanced irradiation effects scaling has been articulated as one of three priority research directions in the nuclear energy part of the Workshop on Science for Energy Technology (Crabtree and Malozemoff 2010). By definition, models developed for materials under neutron-irradiation conditions at this extreme cannot be validated since no/little neutron-irradiation data exists. Consequently, uncertainty quantification will be particularly important in projecting uncertainties into dose regimes that have not been explored using neutrons.

The initiative must strive to be a nationwide network of experts and facilities at universities, industries, and national laboratories led by a lead institution and working as a team. This is clearly a strength of this initiative that was cited by several participants. Challenges include having a well-defined management structure that will promote the partnerships among the participants; having a communications strategy that will ensure frequent and open interactions among the partners; and a central management that ensures that the partners of the initiative embrace the UQ strategy.

The initiative should build a scientifically defendable argument for the applicability of models developed using accelerated experiments to neutron irradiation environments. This should include rate scaling, effects of recoil energy spectra, and the ability to extrapolate to dose regimes not explored by neutrons. The initiative should also design and develop new materials that are radiation tolerant and exhibit smaller uncertainties in in-service properties compared to their conventional counterparts.

To build a scientifically defendable argument for the applicability of models developed using accelerated experiments to neutron irradiation environments, the initiative will need to initially focus on materials where well-characterized neutron irradiated materials and virgin materials of the same pedigree exist. However, there can be uncertainty in neutron irradiation data available in the literature. Further, for LWR materials much work has been done under proprietary conditions and on proprietary variants of a limited number of materials. Much of the data is taken across wide ranges in temperature, flux conditions, maximum fluences, etc. While this may sound attractive, in most cases there was little collaboration between groups to keep the experiments controlled in a systematic manner so that comparison of data across the experiments is difficult. Consequently, the initiative will need to identify the best and most appropriate sources of neutron irradiated material and neutron irradiation data for model validation purposes. Selection of the best material to adopt for the initiative should take advantage of the many decades of experience that exists in the NE community and in Naval Reactors. In one possible approach, the initiative could take advantage of the large knowledge base from LWR materials initially and then move to materials for advanced reactors.

The scope of this initiative is responsive to a request from NE to focus on fuels and cladding. Five topical areas (TA) for research are discussed in this report, each of which exercises different aspects of the methodology. Related work may already be ongoing in DOE by other organizations. The topical areas are designed to demonstrate the added contribution brought by this initiative. These are graded in difficulty and cost. The problems can be sequenced to demonstrate incremental levels of success.

If successful, this initiative will provide a methodology that will enable out of core experimentation to evaluate the performance of fuels and materials. It will also lead to more cost effective and efficient design and development new materials for advanced reactors.

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Crabtree, George, and Alexis Malozemoff. 2010. Science for Energy Technology: Strengthening the Link between Basic Research and Industry. edited by Basic Energy Sciences Advisory Committee. Washington, DC: Department of Energy.

Acknowledgements

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Appendix A: Workshop Agenda

WORKSHOP ON ACCELERATED NUCLEAR ENERGY MATERIALS DEVELOPMENT

MAY 11, 2010

AGENDA

The purpose of the workshop is twofold: (1) to provide feedback on an initiative to use uncertainty quantification to integrate theory, simulation, and modeling with accelerated experimentation to predict the behavior of materials and fuels in an irradiation environment and thereby accelerate the lengthy materials design and qualification process and (2) to provide feedback and refinement to five topical areas to develop predictive models for fuels and cladding and new radiation tolerant materials. It is the goal of the workshop to gather technical feedback with respect to NE's research and development while also identifying and highlighting cross-cutting capability and applicability of the initiative to other federal offices, including NNSA, NRC, BES, FES, and Naval Reactors.

8:30	9:00	Welcome and Introductions	Lesica
9:00	9:45	Plutonium Aging	McLean
9:45	10:00	Break	
10:00	11:30	National Initiative	King
11:30	12:30	Stakeholder discussion	Lesica
12:30	13:30	Lunch and Management Plan	King
13:30	13:45	Introduction to the Topical Areas	King
13:45	14:15	Uncertainty Quantification	Klein
14:15	14:30	Computational Bridge for Nuclear Materials R&D	Bulatov
14:30	14:45	Capabilities and Limitations of Ion Beams	Was
14:45	15:10	Fuels	Pasamehmetoglu
15:10	15:30	Break	
15:30	15:55	Clad materials research	Maloy
15:55	16:20	Fuel/Clad interface	Was
16:20	16:45	Radiation tolerant materials	Allen
16:45	17:15	Summary	King
17:15	18:15	Discussion and action items	All

VENUE

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Agenda date:	May 4, 2010

BY INVITATION ONLY - To register, please email Trina Voelker at voelker4@llnl.gov

Appendix B: Questions and Answers

This section provides an edited transcript of the question-and-answer sessions following each presentation at the May 11 workshop as well as answers to written questions submitted by participants.

Plutonium Aging, Bill McLean

John Hack, Bettis Atomic Power Laboratory

Q: You accelerated the damage by 16 times. Did you also have to accelerate the recovery in order to balance that?

A: Yes, we did. Thank you for asking that. We needed to find a temperature at which we could anneal the damage back out at a rate that was proportional to what we were putting in. We almost got that right. We used theory to guide us there. We stored the samples at elevated temperatures in little boxes that we referred to as “incubators” for the duration of the test to try to compensate. I think the Helium bubbles were just a bit bigger than what we found in naturally aged material.

Cetin Unal, LANL

Q: At NRC, when they deal with probable distribution for best estimate, like metallurgy, they usually take the 95 percentile, like 2 sigma, to deal with the tail. How did you handle that by comparing two distributions you showed for U1 and U2?

A: We basically used the same definition. We actually publish it as either 1, 2, or 3 sigmas because some laboratories, particularly ours, like to report 1 sigma. Other laboratories prefer 2 sigmas, and good scientists prefer 3 sigmas. So we report it all three ways.

National Initiative, Wayne King

Ron Omberg, PNNL

Q: This probably isn’t a fair question at this stage of your program development, but it’s probably useful asking it so you can at least hear it. I’m not a metallurgist. I spent my career on the user end as a core designer. And core design is truly knuckle-dragging engineering compared with what you’re presenting here. But as a core designer, we only wanted two pieces of paper out of the whole metallurgy program. I’m having a hard time imagining those two pieces of paper for this program. Paper #1 is radiation swelling as a function of temperature parameterized with respect to fluence. Paper #2 is radiation creep as a function of temperature parameterized with respect to fluence. So I would suggest, even if you can’t answer the question now, that you think about producing those two pieces of paper.

A: I think that’s a very good suggestion.

Abdellatif Yacout, ANL

C: I think within the fuel presentation, there is one slide addressing the kinetic models for fuel swelling and how it's connection to ion radiation. I think there can be a discussion of this addressing your point about fuel swelling.

Jeremy Busby, ORNL

C: This comment comes from someone with a lot of experience in ions. Concerning slides 9 and 23. I feel like we're selling the neutrons short in both your table and flowchart. I think they're both accurate, but on the neutron side, we're selling things short, like the temperature control. In HFIR and ATR, we can readily control things to the same level, ±10°C. We can do lower dose experiments. For instance, Meimei Li, who's sitting behind me, was doing experiments that lasted a minute or 5 minutes or hours. They're not common, but they're capable. I just want to make sure we're not selling the neutrons short.

Q: Regarding the flowchart about UQ (your slide 19, slide 18 in the book). I'm not a UQ person. When you described this slide, the boxes on the right, the measurable or not measurable. Wouldn't it be ideal to do both, develop a physics-based model and do the experiments? Aren't they both necessary? You sort of implied that it was one or the other: measurable or not measurable.

A: I think all models have to be validated. You have to do validating experiments. That comes when you go back through the loop again.

Roger Stoller, ORNL

Q: The UQ part of this is probably something that's worth thinking about in the context of all of our programs. I first ran into something like this with a colleague at Rolls Royce doing these sorts of things with reactor pressure vessel embrittlement models. The question I have, if you go to the slide where you had vanadium and tantalum. (slide 23?)

The tantalum figure (upper right picture) is the sort of thing that I would expect to see from what I understand of the approach, which is you miss the time and temperature dependence because you're probably missing a mechanism in your model. You look like you're fitting your data, but as you extrapolate, the deviations get larger and larger. I don't see here an appreciation for how in fact changing the rate can actually introduce completely different mechanisms when you cross regime boundaries. That taking a mean-field model, a phase-field model, a Monte Carlo model, whatever you have, what you get out is what you put in there. If you don't have a mechanism in there, you're not going to see a result. Radiation drives the system far from equilibrium, and you're not always going to catch that. So you're uncertainty quantification becomes useless because there are unknowns that you don't know. Moreover, it isn't simply a matter of rate; it's a matter of particle spectrum and particle nature.

I refer you to an experiment that was done at the same dose rate using protons, using neutrons, and using electrons, in which the response of the material, which was pure copper, was completely different. So there's a lot here that's missing or at least that's not being addressed in what I can see.

A: *[King]* In regards to your first point, which was UQ dealing with unknown unknowns, I'd like to ask Tom if he'd like to comment on the model.

[Arsenlis] So this is my work. The point being, in this tantalum model that you're talking about, there actually is a mechanism change in the mobility. So that's the reason we're able to match those two points both at 10^5 and 10^7. We actually do a mechanism flip. The models such as Steinberg-Guinan and PTW and all these other strength models actually used in the code do well in the HE-driven regime, where they're fit. So if the other lines were plotted there, they'd be in the error bound as well. But then as you take them to these higher rates with the laser drive, they don't necessarily have the right kinetics in the phase space, so they fail spectacularly, even though they were experimentally fit to some other regime. So you're not guaranteed that you're going to succeed and fit it.

I think in the second point, let's just talk about modeling now. If you're going to create a robust model, that could in essence give you the microstructure that you saw in all of the three or four types of those radiations, with the same structure, only maybe changing the input that you're driving it with. Then I'd say you have a very robust physics model that you would trust now to another regime. If you don't have such a model, then you basically start from zero every time you're asking a different question about a different radiation condition. I think the point here is to build physics models with understanding, tune them, parameterize the constants as best you can with all the radiation information that you have—albeit potentially accelerated—then project forward and see how relevant that is. That's how you build strength of what we're putting forward.

[King] I'd like to ask Richard to comment on unknown unknowns and missing physics because this is something that UQ takes very seriously.

[Klein] That's a very good question. One of the things that we can do with UQ is discover model inadequacy. Model inadequacy could mean that the models one is using (I don't care what the scientific target area is) are not sufficient in and of themselves to explain the data, vis-à-vis too large uncertainty within the models themselves. If one can determine and start to put bounds on that uncertainty and get that into a comfortable range, but still can't explain the data, that is telling us that there's physics that's not being included in the simulations. This begins to narrow the regime of where to look for that new physics. So that's how one starts to approach unknown unknowns with UQ theory.

[Comments off mic]

[Klein] Well, it's more than just missing the target. It's convincing yourself that you have an adequate quantification of the uncertainties in the models you already have in the problem. If using the approaches and mathematical methodologies, you can convince yourself that the uncertainties that you have are within reason and reduced to the level that you can, then you can really start to look for models or physics that you don't have. It's not a magic bullet, but it starts to narrow down the process and show you … I simply cannot explain …

Let me give you an example: We have in the area of science-based stockpile stewardship, we've brought in all the data we can to basically do UQ on let's say a certain underground nuclear test. We find that we cannot, using all that data and our analysis, explain within reasonable uncertainty bounds the target data that we're going after. That leads us to believe that either there's something wrong with the experimental data that we're bringing in to calibrate or in fact we're in a regime now where we don't have adequate physics.

Rick Kurtz, PNNL

Q: My first question is really essentially the question that Roger asked, which is how you deal with the unknown unknowns. But in looking at the chart about how the uncertainty quantification is to be used, you mentioned you used validation data from reactor or neutron-irradiation experiments. From my experience, that is not a monolithic body of information. There's a lot of uncertainty—or can be—in data that you extract or mine from the literature. I think that's going to introduce a certain amount of variability or uncertainty in interpreting your results.

A: *[King]* In the case of topical area 1, we're going to find the material that has the best pedigree that we can find. And then the PIE is going to be carried out by this team. We're not going to take the results from the literature.

C: OK. But, even if you take that material and characterize it for a very specific set of conditions. If you take that material and put it in another nuclear reactor, you could end up with different results. Frank Garner can speak to these results rather eloquently.

A: *[King]* In fact, just as the uncertainty quantification deals with the model, it also deals with uncertainties in the experimental data. There will be similar things with the ion irradiations, in calculating the dpa, for example; uncertainties in electronic stopping and nuclear stopping. All those things have to be rolled up into the UQ.

C: Your neutron-irradiation database is going to be limited in how far, or to what extent that data dose levels. You won't be able to find 200 or 400 or 600 dpa information on very much that's available to validate your ion-irradiation data. What you can do with ions, as you pointed out, will exceed what you can obtain from neutron archival data.

A: *[King]* But we're going to do something that doesn't exceed that in the first study.

C: You still don't have a mechanism for validating the higher dose information.

A: *[King]* That's right. One of the things it will get you, if you have predictive capability, if you can get to very high dose in some places, you now have a very targeted experiment to do.

Q: The other thing about your description that I didn't quite follow was that you can get to a point where something isn't measurable, I don't understand what you do then. If you can't measure it, how do you ever validate that point.

A: Let's just take the case where a parameter that you need to measure has highly uncertain rate dependence, so that you don't know for sure how to extrapolate that to neutron rates. UQ will give you a hint of that. Then you appeal to a more physics-based model. You go to dislocation dynamics or molecular dynamics or whatever the appropriate model is to address that. Those parameters, which should be more physics based, should be parameters that one has a better chance of being measured. When we go to more physics-based models, this reveals unit mechanisms that we can actually probe.

Q: But at the end of the day, don't you have to have some kind of a measurement to compare against those calculations.

A: You do indeed. All this has to be validated.

Q: If you can't measure something, I don't see how you get to the validation step.

A: *[Turchi, at first off mic]* (Measure … and then with the model, you can …) You may not be able to access one parameter; in the case of fuel, for example, measuring a diffusion parameter may be very tough. But that's not your ultimate goal. Your goal is to explain, for example, site redistribution. So you do your verification on site redistribution, and this parameter you try to get at by modeling.

C: *[Fluss]* The answers that several people gave are correct. The key thing to recognize is that there's a need in this initiative to demonstrate that the methodology in itself is viable. To do that, you must compare ion-beam experiments with corresponding neutron experiments as best one can from existing data or in some cases from data that can be obtained in the near future. Eventually, you will get to the point where ion-beam experiments are working in a regime where similar neutron experiments, if you're using for example dpa's as the scale, are not easily accessible. The ability of the methodology—the bridge, if you will, that comes from the theory, simulation, and modeling—will then have to reach out to a point where direct validation with neutrons might be impossible. However, that doesn't necessarily mean that the phenomenon and the physics aren't revealed at lower dosages, for example. And that will come out of the modeling and the UQ. Comparisons can be made at that level.

So if you imagine applying this to the development of a facility in which materials must last extraordinarily long periods of time—say 50 or 100 years—then you're in a situation that isn't unusual to people in the reactor business, which is a surveillance situation. The methodology that we're talking about could be an extraordinarily valuable tool in a surveillance-type engineering mode, both in the design of the reactor for surveillance and eventually in its maintenance as a surveillance engine. This is used on aircraft; it's used today in reactors. What we're talking about here today is building those tools that allow you to stretch the limits of your comfort, if you will, with regard to operational predictability of the materials and the fuels.

C: *[King]* I'd also like to mention that models could predict ahead, outside the regime of experience. But I just confirmed with Richard that UQ provides us a methodology to predict how uncertainties will grow also as you project ahead. If you do that, you can then ask yourself if the uncertainties are acceptable or not. If they're not acceptable, then you have to come up with a way to do experiments.

John Hack, Bettis Atomic Power Laboratory

Q: While that's slide is up, I want to get back to Roger's point. You said two things about the curve on the upper left. You said that the model was derived with no data, and the other was that you built in somehow a mechanism flip that allowed you to match the eventual data. Was that inspired somehow from something else that said I need to allow that mechanism to operate? Or was it a natural result of some energy balance that the system just went that way and you caught this? Either you had to think about it ahead of time for some reason, or it was just a result.

A: *[Arsenlis]* The way that we found it was through simulation. There's theory looking at dislocation motion in two different regimes—a thermally activated regime and a phonon-drag regime. We conducted simulations to sample the mobility in both of those regimes and then built a function to combine those into a single sort of mobility that spanned those different ranges. In the plots that we get, we actually sample from those different ranges of the mobility and the different mechanisms of that plot. So we actually solved the mechanisms in our lower length-scale simulations.

Q: *[off mic]*

A: It just comes out naturally. If you apply a stress, you get a certain velocity. It has a certain thermal dependence and a certain stress dependence. If you can bridge that divide, …

Q: *[off mic]*

A: So the velocity came from molecular dynamics. Molecular dynamics was used to inform dislocation dynamics. To get molecular dynamics, you use ab initio codes to basically fit potentials. So it was basically all modeling, just passing information along.

Q: *[off mic]*

A: It's hard to obtain that relationship experimentally. It's really hard.

Mike Billone, ANL

Q: Regarding slide for Topical Area 3. Just a couple of comments to help us refine our goals as we go along, and I'll go into more detail in the afternoon. I assume that's supposed to be 100 GWd per metric ton burn-up for the fuel. It was be interesting to study cladding properties up to that limit, but fuel behavior would limit you from ever getting to that point. So you might want to think about fuel behavior during normal performance and accident conditions, which may prevent the cladding from ever getting beyond 70 GWd/metric ton. Just something to think about.

The second point is about the nice picture you have on the left about hardening. We should all realize that the LOCA you refer to is a transient event. In ramping up your temperatures to 1000°C or more, you'll lose all that hardening at about 600. So the picture on the right (hydrogen pickup) is much more relevant to the behavior of the cladding, where you've annealed out cold work, manufacturing and hardening.

A: *[King]* Topical areas 3 and 4 were developed initially in collaboration with Westinghouse.

C: That's fine. I just think we should think this through.

A: You might ask, "Why are you working on LWR fuels? Why aren't you working on more advanced fuels (or materials) for a small modular reactor." That's what we're hoping will come from this discussion.

C: And then a brief comment on Topical area 4. Again, those pictures are excellent, but they're not relevant to LOCA. You might want to clarify what you mean by high burn-up and high power density because in a conventional reactor, you can't have both. The higher the burn-up you go, the lower the power density. Just a few comments as you refine this that you might want to think about.

A: *[Was]* The high burn-up is referring to approaching the 100 GWd/metric ton level. The power density issue is perhaps a linear power issue by continuing to shrink the rod diameters So, we're looking at changes in power density as well as evolution.

Hussein Khalil, ANL

Q: There are a number of facets of this initiative that are all very interesting and useful, but they also seem to overlap very substantially with work that's already going on in DOE programs. There's work on fuels and materials in both the fuel cycle and the reactor development programs. And of course, there's the modeling and simulation activity already going on with goals to develop more predictive capabilities for fuels and materials. There's also a specific activity on uncertainty quantification, verification and validation, and so on. I'm not defending [questioning] the comprehensiveness of these activities. My question is what is the distinguishing feature of this? If you're aspiring, for example, to develop a cladding that can tolerate much more extreme service conditions or something like that, why don't you make that explicitly a goal for this program? If that is your goal—to develop new materials or new models—why not state those goals very explicitly and specifically. Also, I think you should take a look at what is going on already and formulate your ideas in relation in those activities. I would acknowledge that maybe we aren't making enough use of ion-irradiation capabilities and the ability to accumulate damage more quickly, and so on. Why not focus on proposing activities that aren't already being covered in the program?

Q: *[Question to Khalil from Fluss]* What's your opinion about whether that should be one of the highest priorities, namely radiation-tolerant materials?

A: *[Khalil]* No question about it—the materials' performance limits what we can achieve in reactors as far as the capabilities of the system to achieve higher levels of burn-up, to achieve improved economics. There's no question that the incentive is there to develop improved materials. But it doesn't come out as a clear objective of this. Developing, for example, a fast reactor cladding material or structural material that can sustain much higher fluence would be very key to achieving high burn-up and reducing requirements for fuel and recycle.

C: *[Some discussion off mic]*

A: My feeling about this is that we could have picked different problems. But then, would we be picking the most relevant problems? I admit there is intersection with the NE program. This afternoon, we'll ask you to help us avoid any duplication of effort and how best to benefit from leverage on that. There's absolutely no reason why 100% of the work has to be done within this effort. But you asked what distinguishes this initiative. What distinguishes this initiative is uncertainty quantification being used to integrate theory, simulation, and modeling with experiment in the materials enterprise. I think that's unique.

Cetin Unal, LANL

Q: QMU, or UQ, is very important, indeed. However, in the program Hussein was referring to, we are facing the same type of issues. At this point, the multiscale validation methodology is not very clear. This problem we are going to solve from atomistic to meso to engineering scale, tying those simulations together, how you are going to validate them is a challenge. It is not the same problem that we solved in the weapons program. It's a big challenge. For example, in the talk, you made the assumption that top down from engineering scale to meso and atomistic scale, you want to use the UQ that way. I would probably want to do the same thing. However, if you take the FRAPCON code, you may come up with different conclusions, because that code is really an empirical code. We look at that problem, most of the time, when you put uncertainty distribution to parameters, when you run the code it fails, especially with cladding stress calculations. So I think you need to have a good physics model first.

A: *[King]* That's absolutely right. You have to start with the highest level with the best model you can find. This may involve putting new models into FRAPCON.

C: OK. If you say it that way, yes. With FRAPCON, I'm not sure we will have good conclusions.

A: Exactly the same thing happened with multiscale modeling program. The existing codes in the so-called Livermore Blue Book simply did not have the right physics in them. So a new model was developed. That's exactly the approach that we're planning to take.

Carl Beyer (FRAPCON developer), PNNL

C: If you get into the regime where you have very high deformations and necking on the cladding, that's when you'll get some instability. But generally by that time, you're into the failure mode. So you're already predicting failure once you're into the necking mode, especially on irradiated cladding. You almost have nil ductility. There are improvements there, particularly in the deformation area. Deformation of irradiated zircaloy is very complex, and we need better information to model it better. It is, in the deformation area, an area that's modeled on an empirical-type basis.

Hussein Khalil, ANL

Q: I just wanted to follow up on the uncertainty quantification as the organizing principal for the whole thing. My concern is that when it comes to modeling fuels behavior and materials behavior, I think our limitations of understanding of the physics and the phenomena are so great that the unknown unknowns really dominate. I have a difficulty seeing how that can be the integrating theme for this?

Q: *[to Khalil from King]* So do you know that the unknown unknowns dominate, or do you think that they dominate?

A: I'm not an expert on fuels behavior, but

A: *[off mic]*

C: *[King]* So if the unknown unknowns dominate, maybe we shouldn't be doing fuels.

C: *[Ron Omberg, off mic]* Why shy away from a real problem?

A: *[King]* Well, that's why we put it in. We wanted to take problems not that were simple modeling problems, but problems that, if we succeed, would have real impact. The work that was done in the weapons program, you saw it this morning, was a tough problem, not as tough as your problem, but it was a tough problem, and the solution made a real difference. So the question is can we take that same sort of approach, translate it to this field, and pick the right problem where we can have a big impact.

[Fluss] There's one thing that Wayne's diagram may have left out: If there's no starting point, namely, if there's no first-order model on which to build and ask questions with regards to uncertainty, then the first challenge is to build the best first model that you can. Maybe today, without the type of formalism that we're

describing here, that looks like a daunting and maybe impossible path. But with this formalism and recognizing that this process is iterative, maybe it reduces the barrier to saying, “I’ll do the best I can with a first-order model, even a toy model.” Then through a combination of smart experiments and understanding at least pieces of the materials physics, I’ll keep iterating until I have the level of understanding that’s necessary. That’s the sort of philosophical approach that one is suggesting here for the most daunting of the problems, particularly in the fuels area.

Randy Lott, Westinghouse

C: It seems to me that one issue is that it’s important to start with the goals before you choose the models. We’re sort of letting the models define what we can do or what we’re going to do. We had the question when we started was, all I’m really interested in when designing fuel is swelling and creep. Well is that really true or is our goal to have a better alloy for whatever reason or is it to have an engineered fuel-rod design that has perhaps liners or other things that we might do engineering-wise to improve performance. If those are our goals, then what models do we need to meet those goals? Rather than, what models do we have that we can take advantage of to use this great technique? To me then, you are going to be flying all over the place.

Q: We’ve talked a lot about fuels. I have a question about Topical Area 1. It talks here about going after things like yield points, mechanical behavior, but it doesn’t talk much about the goal in terms of there are a lot of different mechanisms and models for radiation damage to different alloys: nickel-based alloys, steels, different kinds of steel. Even two stainless steels have different operative mechanisms and would be a totally different problem. So again, I think we need to define a goal here.

A: *[King]* You’re absolutely right, and let me tell you what we’ve done in that regard. This is an issue that was not resolved before coming to this meeting. The key issue was to find a material that was irradiated in a reactor to high dose where there was virgin material for doing the associated mechanical and ion-irradiation tests. So we’ve looked at materials from Halden, from Bor-60, from a stress corrosion-cracking group that has a variety of materials, and we simply did not settle on the material before coming to this meeting. When proposal is prepared, we intend to identify the material and identify specifically what we propose to do.

C: *[Lott]* Again, I think it’s important not to let the material to define the goal, but to let the goal define the material.

A: *[King]* Regrettably, I think for Topical Area 1, the material may define what we do because of the limited availability of material irradiated to high doses with neutrons. But that’s not going to be true for Topical Area 3, where we pick a particular problem and develop a strength model with radiation creep.

Roger Stoller, ORNL

Q: Your last answer prompts another question. A thing that’s missing here is history. The experiment you’ve described has been done several times in the past. There’s a

20-year program at Oak Ridge involved triple ion-beam irradiations for the breeder program, for the basic energy sciences program, for the fusion program, where all of these rate effects, these composition effects, the helium-to-dpa-ratio effects have been looked at, comparisons made, and the technique found wanting to really be predictive at low-dose neutron radiation conditions. I don't see any mining of this history. It's not just the history at Oak Ridge. The folks at PNNL were involved in this activity; there are ion-irradiation facilities at GE, at Westinghouse, as well as internationally. I feel like there's a lot of acting like something is new here without mining what is well known. If it was a research proposal from a graduate student, I'd tell them to go read the literature.

A: *[King]* You're absolutely right, and I told you at the beginning, this is not a research proposal yet. We're in the definition stage, where we're defining what problems we do. When this group settles and says Topical Area 1 is something you should be focused on, then, we'll mine all of that data.

Ron Omberg, PNNL

C: Someone mentioned that maybe you should concentrate on the cladding and ignore the fuel. That comment sort of came and went. I would think about that. If you go back to FFTF design days, for a while, we tried to separate the fuel behavior from the cladding behavior. But they're two adjacent, interacting systems. Then you put the test into the reactor that has both fuel and cladding, and it responds differently to either one behaving independently. The whole interaction became known as the fuel adjacency effect. Everything you didn't understand became the fuel adjacency effect, which became for a while bigger than anything you did understand. I'd think strongly before dropping the fuel. *[Comment off mic]* If I were you, I'd keep the fuel in there.

A: *[King]* I had similar feelings and worry about the fuel tasks until we visited INL a several weeks ago. Having them on the team gives me a lot more confidence that we'll make progress in the fuels area. You'll see when we give you some timelines later today. Our very first version of the timeline started work on Topical Area 1 and the radiation-tolerant materials and pushed everything else out 3 to 4 years. We've been able to move the fuels area up earlier because of our confidence in working with the INL folks.

Uncertainty Quantification, Richard Klein

Roger Stoller, ORNL

Q: On the contour tree, how do you preserve the information about the gradients?

A: The gradient information is preserved in the distance. These were not arbitrary distances between the points. The distances map directly to the gradients in the actual topo map.

Q: So the length of the green line on the upper right is proportional to the gradient between the two points.

A: Yes, that's correct, and it's also true with all of the other critical points on that map. I should have pointed that out. Thank you for bringing that up.

Moe Khaleel, PNNL

Q: How are you proposing to include model uncertainty? You talked about parameters, but you didn't talk about model uncertainty or missing physics.

A: Let me talk about model uncertainty before we talk about missing physics. Some models have parametric representation; other models do not. I'll give you an example. We do equation-of-state (EOS) calculations, and they use various approximations on the atomic level, like Borne approximation or something like that. Typically, those EOS simulations are usually done by using quantum-level calculations that themselves have some degree of approximation in them to build an EOS table. We regard the EOS table as a model uncertainty. It is a representation of a quantum mechanical simulation that itself has approximations built in. So we generate a number of EOS tables, and those EOS tables are thermodynamically interpolated to get the range of uncertainty in equation of state across a set of conditions. This is the type of thing you'd have to do with any physics model that doesn't have a direct parametric representation.

Q: This gets tricky when you talk about trying to propagate uncertainty in scales. One of the things you think about is what would I pass from scale to scale. I may skip scales.

A: It's a very good question: how do you cascade error across multiple scales? The only way I can see doing it is to do UQ on a given scale with the types of techniques I talked about today. Then you use the output of that—because the output of that is giving you results with uncertainty—as your input with the uncertainty on that input onto the next scale up. So you're now taking that as initial conditions, but initial conditions with uncertainty bounds on the next scale up to propagate that through your models on that scale and bring it up to the next scale. The key is what you do at the interfaces, and I believe the way to handle that problem is basically inputs from previous scale with uncertainties. It's the only way I can understand how to do that.

Q: Are the response surfaces deterministic or stochastic?

A: They are deterministic, but there are errors in them. I've left out a lot of detail here obviously. So the response surfaces are built on mathematical basis functions. You could easily make the statement, how accurate are the basis functions. That's a really good question. Because the accuracy of the basis function determines the accuracy of the response surface, which itself is an approximation of everything else. I have a bullet that says "Advance response surface models." We have dug very deeply into how response surfaces are constructed with basis functions. We've torn apart methodologies like Jerome Friedman's famous multivariate adaptive response lines. We're the first to do this. We're rewriting it in Python so we can parallelize it and get a handle in a very controlled way on swapping in and out different basis function representations to see what the accuracy of that response surface is. That's not been done before. These are very good questions because the methodology itself has uncertainties boiled in.

Carl Beyer, PNNL

Q: In regards to the gradient question, the shorter length means the higher gradient?

A: No, the shorter length means lower gradient.

Computational Bridge, Vasily Bulatov

Unidentified Speaker

Q: You said that UQ is relatively inexpensive, but can you say what level of effort will be spent on UQ? What fraction of the program.

A: What I meant to say is that there are models that are relatively inexpensive that can be used for UQ cycles. UQ itself is not inexpensive.

Q: But it's easily 25% or 30%?

[Discussion off mic]

[Klein] Some of my astrophysics codes take three months to do one 3D simulation on 512 processors going 24/7. There are other codes that can get through something in a fraction of an hour. I don't think you can make a general statement. It depends on the particular piece of physics you're looking at, the level of sophistication in the code and the simulation itself, and how complicated the so-called UQ space is. There could be UQ responses that are very Euclidean in all dimensions, which means you don't really have to do that much to get a good feeling of what the uncertainty is over a large range in domain of parameter space. It's very problem-dependent. I don't think there's a way make that estimate a priori. It's too complex.

[King] Let me mention, we do have an estimate in the budget for this. Remember, we're counting on leveraging a lot of the work that's going on in Richard's group and at your laboratory. The actual number of FTEs is not all that large: 0.5 FTE per technical area, plus a couple that are focused on the UQ methodology itself.

Roger Stoller, ORNL

C: I brought this example up in the meeting in Livermore in the fall. In the days of breeder program, there was a low-swelling austenitic alloy that was developed, tested, and verified based on ion irradiations. Because of phase instabilities that developed under neutron irradiation that didn't develop under ion irradiation, when it was neutron irradiated, it swelled like all the other austenitic alloys. I don't know of any model that can predict that. The complexity of nonequilibrium thermodynamics, which is temperature and displacement-rate driven, is not accounted for by our models.

A: Yes.

C: That's what controls how a material responds to neutron radiation. So there's a big caveat that needs to be embedded in all of this.

A: Yes.

Luke Brewer, SNL

Q: So you showed the dislocation dynamics simulations based on the distribution of interstitial loops, and you had different densities of defects. If you were to redo that simulation at the same nominal density of defects and just randomly move them around, at what point … those are all deterministic simulations, correct? Have you gone back and checked … you have the uncertainty of your results at any specific length and time scale. At some point, the different between 10^{13} and 5×10^{14} doesn't matter because if I do the simulations several times, I see that the results completely overlap.

A: *[Arsenlis]* So this is my simulation. The way we typically run these is we try to run in large enough simulation volumes that allow us to develop the stress–strain curves—you can see the level of noise that's in them. It's very low for a simulation of that detail. With regards to the initial microstructure, the initial loops are all placed randomly. It's an instantiation of a random microstructure. Now the question is, Would a different random microstructure give something different. I think on a small volume, if you didn't have a large computer, you'd have to do such an exercise and do basically replicas to average out the noise. In this case, we're able to run on a third of BlueGene for three months, large enough system so we can sample all those replicas simultaneously. I think we address that problem by running big.

Q: *[off mic]*

A: The strain rates on the order of 1.

Ion Beam Capabilities and Limitations, Gary Was

Carl Beyer, PNNL

Q: I'm interested in your slide that showed crack length vs. weighted average channel height. The weighted channel height is dislocation channel?

A: Yes.

Q: We have a similar issue with zircaloy as well, and we haven't done enough work to correlate stress-corrosion cracking with dislocation channels. We do see very localized deformation, and dislocation channels have been observed. I believe we're seeing something similar with zircaloy as well.

A: This is for stainless steels. In this database, there are seven alloys, two doses, two strains, one environment. I didn't distinguish between them because what I'm trying to highlight is the degree of localized deformation, which is driving irradiation assisted stress corrosion cracking.

Q: Right. And the red circles and squares?

A: Anything that's open indicates no cracking; anything that's solid indicates cracking. Basically there's a narrow threshold or window.

Q: Are those different materials?

A: They're all stainless alloys, but with different chrome and nickel compositions.

Nuclear Fuels, Kemal Pasamehmetoglu

Mike Fluss, LLNL

C: I'd like to clarify what I believe is how one would use heavy ions, ions that look like fission fragments, in studying fuels. Once you have a heavy ion that is in the vicinity of a fission fragment energy, you can look at both the nuclear stopping, which accounts for about 2% of the energy and results in displacements, as well as the electronic energy loss. What that does was a subject of discussion. Wirth and Olander had an interesting presentation on this topic—I don't know if there was a corresponding publication—so-called Coulomb explosions in insulator or ceramic-type fuels. Nevertheless, a large fraction of the fission energy (98% of it) is deposited as electronic energy loss. So ion beams in the range of 100 MeV that simulate fission fragments are available at a small number of accelerators around the country and worldwide. There is a similarity that's achievable in the laboratory. But there are many applications as you pointed out. One can inject fission gases. One can put markers in and then look at the diffusive properties of the markers. One can establish let's call them simulants of different burn-ups and then drive those systems with the ions and look at productions of second phases. The point I'm trying to make is, it's a quite flexible platform. However, you probably can't do all of those things at the same time. That's both the good news and the bad news. The bad news is if you do all of them at the same time in a reactor, you get what you get. In a sense, if you're looking for something that looks like separating different mechanisms experimentally, the ion-beam platform provides you that access.

A: I agree with that. But as you know, there's a synergy between both mechanisms. If you only rely on one, and then say that the model I developed for *this* is going to directly extrapolate to *that*, it's not going to work.

C: That's the reason I hate the word separable variables, because they are hardly ever separable. It helped to study them separately.

A: Yes, it helped to study them separately, but it also gives you the added advantage of identifying that synergy when you compare the two data together.

Vasily Bulatov, LLNL

C: You said (in my own words), only models that reach some level of maturity or reliability deserve the UQ treatment. In my understanding of what UQ does, it's probably the most reliable way to do model validation. I don't quite understand, who would establish that a model is mature unless we quantify and validate it?

A: When I referred to models, I was referring to the fundamentally based or mechanistically based models as opposed to empirical models that the current codes have. I do worry about using UQ, especially using sensitivity analysis, on empirically based models. Even though the absolute magnitudes within a certain range may not be too bad, I really worry about the sensitivity and the partial derivatives of those empirical correlations. That's when you start getting into trouble, and they start pointing in the wrong direction.

Clad (Core) Materials Research, Stuart Maloy

Dave Gelles, PNNL

C: We did some experiments a long time ago that involved neutron irradiating materials and then electron irradiating them. We found that there's a temperature shift that you have to be aware of. If you don't take that into account in these experiments you're planning in the ferritics, I anticipate that you'll have confusing results.

A: That's an important point, and Gary has pointed it out. We've done that on some of the ion irradiations we've done on other ferritic martensitic steels looking at hardening, in a sense, versus dose, and you can see that in those specific materials. We'd have to do the same thing if we're going to push things to higher dose, we'd have to look at that temperature compensation.

Fuel–Clad Interaction, Gary Was

Carl Beyer, PNNL

C: I have two comments. First, about your idea that power upgrades will lead to more PCI problems, actually that has already been seen. There seems to be a strong correlation between those BWR PCI failures in those plants that have already had upgrades. We're already seeing that.

On slide 10, the different models involved in gap closure. There's another one, particularly when you have a power increase, which is usually the mechanism that's driving the PCI. Once you get above a certain burn-up level (about 40 to 50 GWd/ton), you start to see gaseous [fuel] swelling as well. That can add additional stress on the cladding that's driving fuel out. In LWRs, we don't see it until you get to 40 or 50 GWD/ton but it seems to be pretty strong once you beyond those burn-up levels.

A: Yes, that's not on here. This is pretty linear as you can see. It's solid fission products. That's another one that doesn't even show up here. When that takes off, it takes off highly nonlinearly. Very good point. Since it increases so quickly, it can drive the uncertainty in when this closure occurs very dramatically.

C: And the uncertainty in the cladding strain that results, so it's a big uncertainty that codes have a difficult problem in predicting.

Mike Billone, ANL

C: Let me kill you with kindness first. That's an excellent presentation on a difficult subject. If you could survive all of this and get to maybe 40 to 50 GWd/metric ton, you want to also add in that you have a fuel-cladding bond oxide layer on the inside of the cladding that may protect you from some of this chemical interaction. You also start to get that fuel rim, which is fairly soft and high in fission gas bubbles. Maybe you can survive. It's different for a BWR and PWR. For a PWR, you start forming that bond at 30 to 40 GWd/metric tons. High burn-up fuel may be more benign if you can make it through this stage. A lot of the mechanisms that you show are very important for lower burn-up fuel before you form the fuel-cladding bond and you get the soft rim. If you could get through that critical period, there are high burn-up facts that may help you out.

A: Good point. Thank you.

Carl Beyer, PNNL

C: A lot of the PCI failures are happening in the second cycle. They're around 30 to 40 GWdays/ton. We also don't know what the mechanical strength of that fuel-clad bond is. It can act like a liner. If it's fairly ductile, it could improve the PCI problem. If it has more strength, it may enhance the PCI problem. That's an issue. It's probably softer than the UO_2 is because it's a mixture of zirc, uranium, and oxygen. That's a real complicating factor.

Radiation-Tolerant Materials, Todd Allen

Dave Gelles, PNNL

C: Let me add another level of complexity to your problem. Once you've defined the material, you still have to make product form that can be put to use. The example I give you is MA957, which you've heard of. We took a couple of years to learn how to make tubing out of it and another couple of years to get vendors to produce the tubing. When we got the tubing back, it was cracked. Then we ran out of money.

A: *[King]* I want to make a comment on that. Is the QuesTek representative here? I think you often take that into careful consideration—the fabrication of the material. Isn't that correct? Sometimes you'll sacrifice properties for fabricability in the design process

[QuesTek rep] Yes. *[Other comments off mic]* . *[Olson]* The inefficiency of traditional empirical materials development (whether model-assisted or not) is well documented. In contrast, the systems-based integrated materials design/AIM

(Accelerated Insertion of Materials) methodology at QuesTek incorporates "design for manufacturability" up front. The principal concept of the AIM component of the method is application of predictive science to process scaleup, component-level process optimization, and forecast of manufacturing reliability. The successful application to the flight qualification of Ferrium S53 at both the material and component level encountered no process scaleup or fabricability problems at the level of full landing gear manufacture. The demonstrated reduction in technical risk attests to the power of experiment-assisted theoretical design as a radical alternative to model-assisted superficial empiricism.

[Allen] But it's a very good point. I talk about modifying grain boundary structures, and we do that through a combination of deformation and heat treatment. But it's got to be consistent with final product form if you want to use that as one of your methods. So it's a very good point.

Vasily Bulatov, LLNL

Q: In your study of helium agglomeration at the grain boundaries, was there any correlation seen between the types of the boundaries, say high end and low end, in terms of the propensity to agglomerate helium?

A: In the older studies I showed you, I don't think they even thought about it at that time. In the newer ones, I don't know that it's been done for this specific system. It's one of the things we'd like to do if we can look at the samples that have been irradiated. And we have some other ones at ATR that will come out. It's definitely one of the things we want to look to understand those correlations. Because if there is no correlation, then it's a quick example of that's not the cause of the improvements in properties. It's the precipitates or some other feature.

Rick Kurtz, PNNL

C: We have lots and lots of specimens that have used that injection foil technique. If you want to take some, we'll give you some.

C: Actually, we have some specimens coming out of HFIR that have over 1000 ppm of helium in a variety of materials, ferritic steels as well as ODS materials.

A: Good point. This could be one of those cases where you don't even need to do the irradiation studies because it's already been done.

C: There's quite a bit already, at least, at modest doses: tens of dpa's. *[Other comment off mic]* We've also been working on developing models to describe the helium transport and fate; this has been going on in the fusion program.

A: So those would be a good modeling start.

Summary and Timeline, Wayne King

Roger Stoller, ORNL

Q: What do you have at the end of 5 or 6 years on your radiation-resistant material?

A: A material that meets the goal.

Q: Which the goal is?

A: That's the problem. We were going to set that here. Right now, the goal, in my mind, is poorly specified. It says only "to develop radiation-tolerant materials and materials that have a smaller spread in properties." We need a much more sharply defined goal.

C: Based on experience back to the 70s, that's a very imaginative goal in 6 years.

A: It is, indeed.

Unidentified Speaker

Q: *[starts off mic]* … in the short term, … modeling effort to look at a post-irradiation property and then coming up with a radiation-tolerant material before you do the cladding work that gives you the in-pile deformation. It seems out of phase. You don't really understand yet how to couple all the radiation-damage mechanisms and their impact on properties, and you're starting to design a material that's radiation tolerant. Seems like that should be the ultimate goal.

A: We considered having the work on the radiation-tolerant material at the end of the process, after we'd done a lot of the other model development. But we also realized the potential to leverage that working in this area brings to the project. There's a lot of interest right now in radiation-tolerant materials. There may be good reasons to try to push that development forward.

Moe Khaleel, PNNL

Q: Have you looked at the NE milestones? Kemal talked about the four objectives, and behind these are a lot of milestones that NE is trying to target over the next 10 years. Have you tried to map the milestones and the activities out of the five focus areas back onto NE's goals so one can see that this activity will contribute to that goal in such and such area?

A: We haven't done that yet, but it's something we should do. We took NE's instructions to look at fuels and clad. We asked them after our March meeting if we should present these five topical areas at this meeting. The answer was yes. We've phased them based on our best guess at how we can put human resources and physical resources behind the problems.

Q: Do I understand it correctly, if I asked you about priorities for the five focus areas, based on the timetable that you presented, that your priorities are focus area 1, 2, and 3?

A: I would say I have the highest confidence that we can do topical areas 1, 2, and 5: the model development, the fuels, and the radiation-tolerant materials. There's high impact if we're successful in the radiation-tolerant materials. There's motivation for starting that earlier rather than later. *[Question off mic.]* Yes, 1, 2, and 5 were started early; 3 and 4 were started later—in the phased approach.

Mike Billone, ANL

C: I made a comment earlier in the day that you might want to exclude the LOCA temperatures, the high temperatures, because it didn't fit with the modeling and experimental approaches. Meaning temperatures in which you've annealed out all the temperatures that you're trying to model and annealed out the fabrication variables as you ramp up to 1000 or 1200°C. At the same time, LOCA is a design-basis accident, so somewhere within NE, maybe at the fuel-cycle research and development level, one must consider the effects of going to high burn-up on behavior during a loss of coolant accident where you're likely to balloon and burst the cladding. At burn-ups higher than 70 GWd/metric ton, particularly 90, which has been demonstrated experimentally, when you balloon and burst the pressurized rod, you may be blowing out half the fuel in the rod for very high burn-up fuel. So I'm not suggesting that you include it in your work, but I am suggesting that it's a very important area and, at some level, needs to be considered within NE.

A: *[King]* That's a good comment on the LOCAs. We'll certainly take that very seriously.

Mike Fluss, LLNL

C: Since one of the goals that Sue mentioned this morning was breaking down the cylinders of excellence, as I've heard it called, and since we have the principal investigator on a very relevant Energy Frontier Research Center (EFRC) project, I was wondering if I could ask Mike to say a couple of words about how he might see his EFRC interfacing—intellectually, not monetarily—with what has been proposed here. our proposal?

Mike Nastasi, LANL

C: Actually, there are three EFRCs that have overlap with what's proposed, especially in developing radiation-tolerant materials. My EFRC specifically is looking at the role of how interfaces influence materials in extreme environments—both the radiation and mechanical extremes. So half of my EFRC is definitely aligned with looking at radiation-tolerant materials.

Post-Workshop Questions and Answers

The following questions were submitted by workshop participants following the workshop and were answered by members of the initiative team.

John Hack, Bettis Atomic Power Laboratory

Q: In the work of Arsenlis on V, a model was built without data but caught a flip in mechanism that made the model predictive at high rates. Was that mechanism a natural result or built in a priori (and why)?

A: *[Arsenlis]* The mechanism flip is known to happen in a wide variety of materials, but the exact shape of the dislocation mobility curves, and the stress/velocity levels at which these transitions occur is not known a priori. Thus molecular dynamics simulation or experimental measurement is required to obtain the exact nature of each specific material.

Roger Stoller, ORNL

Q: Ion irradiation failed spectacularly to predict the neutron-induced swelling of LS-1, an austenitic stainless steel developed during the breeder program. Nonequilibrium conditions are strongly rate-dependent and not predictable by current models. Simple temperature shifts don't work. [Singh, B. N., M. Eldrup, K. Horsewell, P. Ehrhart, and F. Dworschak. 2000. On recoil energy dependent void swelling in pure copper—Part I. Experimental results. *Philosophical Magazine a-Physics of Condensed Matter Structure Defects and Mechanical Properties* **80**(11), 2629–2650.]

A: *[Arsenlis]* I believe that this is a great result. The modeling challenge is to determine what the source of these differences is and incorporate that in to a robust model of microstructural evolution and macroscopic response capable of explaining these discrepancies. I believe that such a program is being promoted here.

Luke Brewer, SNL

Q: With the dislocation dynamics simulations (Bulatov, Slide 14), if you do Ta simulations with multiple instances of the microstructure, what will the distribution of stress–strain curves look like?

A: *[Arsenlis]* This depends on the size of the simulation cell that is used and the density of dislocations within. For low densities with small simulation cells, the difference between different simulations instances can vary from one to the other. With high dislocation densities, and large simulation cell running on high-performance computing platforms the simulation-to-simulation differences disappear as the densities and number of objects become statistically significant.

Roger Stoller, ORNL

Q: PKA energy spectrum effects can be just as significant as rate effects. See, for example, Singh et al. comparison of swelling of pure copper under neutron, proton, and ion irradiation.

A: *[Fluss]* While dpa scaling is often applicable (compensating for rate effects) it is important to consider the consequences of PKA energy spectrum effects as well as alloy compositional change and radiation enhanced diffusion to mention just a few. Accelerated experiments should be designed in such a way as to explore such questions rather than to ignore them.

David Gelles, PNNL

C: Let me add another level of complexity to your problem. After you have chosen a new product (alloy), you then need to manufacture it into a useful product form. An example: MA957 for cladding had to be manufactured into tubing. It took us two years to learn how to make tubing, another two years to teach two vendors how to make tubes, and when we got them back, we found they were cracked, and we ran out of money.

A: *[Fluss]* Ones ability to fabricate a particular material into a useful technological component cannot be under estimated when considering the time scale for the successful insertion of new materials into nuclear energy systems. What this initiative offers is the possibility of getting to this stage in the development of a material earlier than if one had to wait for the results of experiments that take much longer times.

Jeremy Busby, ORNL

Q: Regarding Campaign 5, you want to use UQ for alloy development? How will that work? We already make use of advanced computational tools for alloy development. Specifically, we use computational thermodynamics to optimize alloys well ahead of actual melting. It's not clear how UQ applies here. Alloy development is also being pursued in the Fuel Cycle Research and Development (FCRD) Program, Gen IV (SFR [Solid Fuel Reactor], NGNP [Next-Generation Nuclear Plant], and others), and LWRs in the coming year. In addition, we hope there will be an open competition for blue-sky alloys as part of the Nuclear Energy Enabling Technologies (NEET) Program in FY11.

A: *[Fluss]* UQ guides engineers, experimentalists, and modelers to focus on the highest leverage properties of a material under development, those properties which are characterized by high uncertainty and high sensitivity with respect to performance. Gilding the lily is not what we want for nuclear energy materials development.

[Olson, QuesTek] UQ plays a central role in the computational materials design approach developed by QuesTek, supporting sensitivity analysis for robust design of material composition and processing. It also supports a "design for manufacturability" approach that anticipates the full range of materials processing requirements. Anticipation of process scale in early design supported the demonstration of full flight qualification of the Ferrium S53 stainless landing gear steel with no scaleup problems.

C: Further, regarding UQ for alloy development, you've stated that you expect an alloy to be ready in 6 years. There is some concern with this claim. It demonstrates a lack of understanding about alloy development for reactor applications. There are several required tests for ASME qualification and (hence) NRC approval that will run 5-6 years by themselves. There is a considerable amount of testing required beyond the irradiation experiments that will be required. You might have a concept that has been validated in 6 years, but it will not be ready to go into production.

For comparison, in SFR and FCRD, it will take us about 1–2 years to get a candidate alloy that's been optimized using modern computational and materials science methods. Then, in parallel, we're also working on identifying any fatal flaws (irradiation, corrosion, welding, creep, tensile, fatigue, etc.) After 5-6 years, we plan to be at a spot where we can state with some certainty that the alloy will work. It will then take another ~10 years of data collection to get it qualified. It's not hand it off to a vendor in 6 years for instant production.

A: *[Fluss]* Reducing the time scale for the front end of the value chain does not address required tests. We are hopeful but not certain that by working with NRC at the earliest stages of this initiative we may be able to use some of the same principles to help reduce the back-end of the materials development and insertion process. This will not be easy but if something like this is to ever happen something like UQ will have to be employed.

[Olson] The application of UQ in the computation-based acceleration of the full materials design and qualification cycle was the central concept of the DARPA-AIM methodology, as highlighted in the 2004 NRC report *Accelerating Technology Transition*. The AIM project demonstrated successful probabilistic modeling of microstructure variation over six stages of aeroturbine disc manufacturing, enabling accurate forecast of 1% minimum mechanical properties for "A basis" design allowables. QuesTek has now applied the methodology to the flight qualification of the Ferrium S53 stainless landing gear steel, meeting all mechanical property requirements while demonstrating the successful prediction of the 1% minimum strength levels necessary for the MMPDS (Metallic Materials Properties Development and Standardization) design allowables. The fully computationally designed alloy went from a "clean sheet" to flight qualification in 10 years, getting everything right up front through full use of predictive science. Based on the technical path followed, it is estimated that the full cycle could have been completed in 6 years if funding had been continuous. This cycle is the goal of a second landing gear steel currently entering the qualification phase.

C: Slides 8 and 23. I strongly support the ions. But I think we're selling the neutrons short. We can actively control neutron experiments to ±5°C in HFIR, ATR, etc., for example.

A: *[King]* It was not my intent to sell neutrons short. Neutrons irradiations are key to the success of this initiative. I will incorporate the change that was suggested into slides 8 and 23.

Q: On your UQ flowchart, you imply that either experiment or modeling be done (decision part on right). Why not both? Would that be preferred?

A: *[King]* The questioner is referring to a point in the process when a decision is made as to whether an experiment to measure a particular parameter is possible or not. An example of this is the dislocation mobility. Measurement of dislocation mobility is challenging at best. In this case, the decision could be made to appeal to dislocation dynamics for determination of that parameter. When passing through the UQ loop again, validation experiments are part of the process.

John Hack, Bettis Atomic Power Laboratory

Q: It seems out of order to work on development of an "irradiation-tolerant" material before the models for irradiation damage/performance relationships are in place.

A: *[King]* We considered having the work on the radiation-tolerant material at the end of the process, after we'd done a lot of the other model development. But we also realized the potential to leverage that working in this area brings to the project. There's a lot of interest right now in radiation-tolerant materials. There may be good reasons to try to push that development forward.

Ron Omberg, PNNL

C: Suggest that an ASTM-like or ASME-like working group is needed to review and sign off on data qualified for further use by reaction designers.

A: *[King]* Peer review is a critical part of the UQ process. An Expert Peer Review committee is included in the organization of the initiative. Membership on the NE Expert Peer Review Committee is determined by the nature of the particular peer review and, to the extent possible, involves all relevant experts. The premise is that once the community of experts in a given problem area is convinced that the demonstration has been effected, the problem may be considered solved, in a robust and final way.

Roger Stoller, ORNL

C: Proposal fails to acknowledge years of previous experience in U.S. breeder, fusion, and BES programs using ion irradiation and comparing ion with neutron irradiations.

A: *[King]* The initiative has not yet reached the point of becoming a proposal. We are still in the definition phase. When the team is asked for a proposal, they will take full advantage of the previous experience in U.S. breeder, fusion, and BES programs using ion irradiation and comparing ion with neutron irradiations

Ron Omberg, PNNL

Q: How do you get two fundamental outputs: irradiation swelling versus fluence and temperature, and irradiation creep versus fluence and temperature. These were the two fundamental inputs for FFTF core design from the material science people. I can supply examples if needed.

A: Developing models that can predict the evolution of microstructure and properties with irradiation is the goal of the initiative. We will consider both of these suggestions in the planning process for future work.

Michael Billone, ANL

C: On TAs 3 and 4, I recommend that LOCA temperatures and conditions be **excluded** from this program because most of the mechanisms that will be modeled and investigated experimentally will be annealed out. However, LOCA is a design-basis accident and needs to be addressed by DOE NE, perhaps in the FCRD Advanced Fuels Campaign. For example, LWR cladding balloons and ruptures during the LOCA heating ramp at 700º–850°C. The rod depressurizes and blows some of the fuel out of the rod. Up to 70 GWd/MTU, the fuel expulsion is mild. At 90 GWd/MTU, about half the fuel in a Halden test rod was expelled through the rupture opening.

A: *[King]* LOCA will be removed from TA3 and TA4.

C: Correct typo MWd/MTU → GWd/MTU, separate 290–360°C steady state from LOCA 300°C → 1000°C transient. Irradiation-induced hardening, cold-work, and other fabrication variables are all annealed out during temperature ramp at about 600°C.

A: *[King]* Correction noted.

Ron Omberg, PNNL

C: Suggest not dropping fuel behavior in terms of studying only cladding behavior.

A: *[King]* We have decided to retain the TA2 on fuels in the initiative

Jeremy Busby, ORNL

C: I'm still uncertain about the dramatic shift from ion beams as a tool to a UQ mission with ion beams in a supporting role. That movement was so sudden and radical.

A: *[King]* We have been considering UQ as a guiding principle since October 2009 when we were preparing a poster for Secretary Chu's visit. It was adopted into the initiative in December 2009.

C: As I noted at the meeting, I think you're overselling the differences between ions and neutrons. Specifically, I think you've sold neutrons short (and I am very passionate about ions; I spent 10 years using the exclusively). Most folks familiar with irradiations will likely point out some discrepancies in your slides. Several subpoints:

- Energy: can be tailored in reactor. At HFIR, we can harden or soften the spectrum as needed with capsule design
- Products: What do you mean by separable? And, aren't the ion beam activation products also nuclear physics controlled?

- Temperature: Should be temperature control. And, in reactors, we can control them to ±2 degrees in active experiments and in HFIR, we can hit them to ±10°C in our passively controlled capsules
- In-situ observation: Halden can do a lot with in situ testing.
- Unit mechanisms: what do you mean here? I'd say that the difficulties (challenges) for ions and neutrons are about equivalent. Different, but equivalent in magnitude
- Cost: Again, I think you're underselling neutrons. In HFIR, we can do a capsule of tensile specimens (designed, built, irradiated to a few dpa, shipped, and tested etc.) for less than $10–15k. Instrumented capsules are much higher to be sure, but all experiments are prohibitively expensive. Just like not all ion experiments are dirt cheap.
- Time: I assume you mean for 1 dpa irradiation? If not, most reactors also have rabbit tubes that can do irradiations on the order of seconds to hours to days.
- In situ: as above, see Halden's work. It's indeed difficult, but not impossible. Also, in some techniques, small specimens lead to artifacts due to surface effects. This should be captured in later slides.
- Sample thickness: we irradiate TEM samples in reactors all the time at 100 μm.

A: *[King]* Energy: True, but the extent to which this can be done is small. The value is not clear as we really don't know how small differences in spectrum affect the damage state.

Products: At ion energies where nuclear reactions do not occur, the in-growth of fission products or transmutants can be studied individually using multiple ion beams.

Temperature: noted.

In situ observation: There are a lot of problems with these tests, and to our knowledge, they haven't been very successful and haven't yielded much, though Halden has been trying hard for years.

Unit mechanisms: Ion beams have played and will play an important role in discovering the unit mechanisms that are key to materials performance. It is unlikely that radiation-induced-segregation, and its underlying nonequilibrium diffusion mechanism (Okamoto, 1974), would have been correctly identified, even as of today, if only neutron-irradiation effects had been studied. Other such examples include, but are not limited to, the importance of the primary recoil spectrum to freely-migrating defect production (Averback, 1978; Rehn, 1984), the contribution to enhanced diffusion from atomic replacements (Rehn, 1987), nascent cascade development (Jenkins, 2000), and the void lattice (Loomis, 1975).

Cost: Full cost must include the cost of extracting the data afterword.

Time: On a dpa basis, there is still a factor of 100–1000 difference.

In situ: correct

Sample Thickness: Thicker samples for neutron application noted in the table was included as a positive point for neutron irradiation that is hard to replicate with ions

Carl Beyer, PNNL

C: Strength as a function irradiation for most materials used for current LWR are well known (modeled empirically) with relatively small uncertainties. Plastic deformation and fracture mechanisms are not well defined with large uncertainty for irradiated materials with hydrides or voids/bubbles. Cladding failure strains both on a micro- and macroscale vary with material, fluence, hydrides, voids, bubbles.

Regarding UQ, the time scale translation for uncertainty quantification is a potential major problem because [there are] no neutron data for long time periods. Someone pointed out cross-correlated parameters.

A: *[Klein]* Data for long time scales is important. If no data exists at long time scales, then UQ is used to extrapolate uncertainty into long time scale regimes and this will be reasonable if no new physics comes into play at these longer timescales. This is all that one can do.

Jeremy Busby, ORNL

Q: There is also the issue of "knowing the unknown unknowns" that came up during the workshop.

A: *[Klein]* See the answer to the following question.

Roger Stoller, ORNL

Q: Issue of unknown unknowns on UQ and extrapolatability.

A: *[Klein]* Non-intrusive UQ using an ensemble-based approach attempts to take into account all sources of input error associated with the simulations as well as error associated with the measurement observations. The categories of uncertainty or error include error in the observational measurements, prediction error or uncertainty associated with the statistical emulator (response surface or meta-model) both for current conditions where we have observations and for the new conditions where we might not have any observations and the uncertainty in the simulation code unexplained by the parametric uncertainty we have included for the models. This is frequently referred to as the unknown unknown (aka model error or model inadequacy). How can we get a quantitative understanding of the so-called unknown unknowns? The approach that UQ takes is to simultaneously account for (1) observational error, (2) emulation error, and (3) model inadequacy error. The reason being that in most cases one does not have the necessary volume of observations or simulations to characterize each of these terms separately. Hence, given the observations we have and a fixed number of simulations, one can aim to characterize the joint impact of all these sources of error of both UQ of the parameters in the models and the prediction of the state of the system for the new regime we are aiming for. By carrying out a large ensemble of simulations and developing response surfaces and then comparing the response surface predictions with random sets additional simulations that were not used in the construction of the response surface, we can both quantify and reduce the emulation error term. We might also have a good characterization of the observation error term. The model error (unknown unknowns) is what remains; the difference between the simulations and the observations that we cannot explain by observation and emulation uncertainty. As we made assumptions about the statistical properties of the observation errors and the response model, we make a prior statistical assumption about the model error and by working simultaneously with all three sources of errors, we get a quantitative idea about the contribution of unknown unknowns (i.e., the model error term).

Cetin Unal, LANL

C: UQ is important. However, it requires a good description of physics model and characterization of uncertainties. Top-down UQ may end up with wrong conclusion if the top model is very empirical. They crash at certain conditions. So which approach, top-down or down-up, [works] better when you don't have physics. Some examples that Arsenlis explained suggest down to top, finding a new mechanism predicting data.

A: *[Klein]* Physics models need to be verified and validated before UQ is applied. This is not the job of UQ, but of verification and validation (V&V). If physics models have undergone a certain amount of validation in regimes we have data for, than assuming we can parameterize the model in some fashion and characterize the uncertainties, we can begin to apply UQ. Of course one needs to characterize the uncertainties in the model. This is done by collaboration of UQ specialists with model specialists. For full system, a bottom up approach to UQ for the full system is how it is done. This also applies to V&V. The component physics is first verified and validated and then this is aggregated (coupled) to the next component level and all the way up to a full system. Same approach applies to UQ. You start with UQ for the first part of a system and the uncertainties are then fed into the next part of the system in a systematic way. If you "don't have physics" then you go with highly empirical models, but accept that the uncertainties in the outcomes will be possibly very large.

Q: NRC uses 95 percentile value when it deals with best estimated distributions in CASU (Contextual Assessment of Systems Usability) methodology. How did you treat tails in U_1 and U_2?

A: *[Klein]* One experiments with the sensitivity of the assumptions. Typically, we look at 5–95% confidence levels, but we also experiment with going out beyond 2 sigma to see how the UQ outcome would change.

Jeremy Busby, ORNL

C: I fully admit that I'm not a UQ expert. I learned a fair bit from the presentations. But, I still have lots of questions. I'm still uncertain about the stated preference for modeling over fundamental and supporting data. I can see that in some circles (such as your climate example), there are values or constants that are not obtainable. But, I struggle to find examples of that in the nuclear world. As a result, it is tough to see how this can then sold as a valued tool to NRC, ASME, or any other type of sanctioning body that has traditionally been so heavily reliant on tangible and verifiable data. I'm happy to be educated here.

A: *[Klein]* There is not a stated preference for modeling over fundamental data. Both are crucial and go hand-in-hand. UQ would not be needed if we lived in a perfect world where we had access to any experiment we wanted to perform and for any design we entertained and we had truly reliable data for any and all instances we need. Unfortunately, none of the above is true. Advanced modeling combined with UQ analysis is crucial to the design of new reactor concepts and for exploration of new ideas. UQ on models will indicate to us what the large-scale uncertainties are and will suggest experiments that need to be performed to get a better handle on those uncertainties. This will inform our decisions on what experiments to perform and potentially save vast amounts of dollars in a field where experiments are so expensive.

John Hack, Bettis Atomic Power Laboratory

Q: The damage rate was accelerated by 16 in "spiked" materials. Was an adjustment made in the recovery rate to keep the two commensurate?

A: *[McLean]* Yes, we did. Thank you for asking that. We needed to find a temperature at which we could anneal the damage back out at a rate that was proportional to what we were putting in. We almost got that right. We used theory to guide us there. We stored the samples at elevated temperatures in little boxes that we referred to as "incubators" for the duration of the test to try to compensate. I think the Helium bubbles were just a bit bigger than what we found in naturally aged material.

Valery Bulatov, LLNL

Q: Was there any correlation of He agglomeration with the type of grain boundaries?

A: *[Was]* I don't know the answer to this question. I'm not sure I've seen measurements of He association with grain boundaries by type.

Michael Billone, ANL

C: There are two characteristics of high-burn-up fuel that should be added to the picture: Fuel–cladding bond (8–12 μm), which is a $(Zr,U)O_2$, and which may protect the cladding from IASCC. A sub-micron fuel rim, high in Pu and fission gases, develops. This soft rim may mitigate pellet-clad interface (PCI). Thus, if a fuel element can survive two fuel cycles (≤45 GWd/MTU), it has better odds of surviving a third and fourth cycle.

A: *[Was]* Good comment.

Michael Billone, ANL

C: For conventional LWR fuels, high-burn-up and high power density are inconsistent.

A: *[Was]* Not really. High power density refers to power uprates. Higher burn-up refers to longer life of fuel, so you can have both. There was another comment later on tin which it was pointed out that power uprates have already led to higher failure rates.

Unidentified Writer

Q: Microdeformation is different between non-irradiated versus irradiated zirconium alloys; dislocation channeling may be an important mechanism.

A: *[Was]* Correct.

Jeremy Busby, ORNL

Q: On a more specific note, you frequently call out life extension issues for LWRs. That's a key element of the NE program. But, fuels are not a limiting factor for 80 years. The fuel is frequently replaced. While fuel reliability is an economic factor and under the broadest definition of sustainability, this is not a life-limiting factor at all. It should be tied to structural materials if you want to look at limiting conditions for life extension. Moreover, if we get beyond that point, why focus on SCC of clad? That's responsible for virtually no fuel failures. Industry isn't all that concerned with it. So, why are we? Wear, fretting, debris are where the real action is at.

A: *[Was]* Correct. Fuel behavior is not a life extension issue. This comment is correct – which is why I focused my talk on fuel failures more broadly than just SCC, which is still an issue for BWRs but not much for PWRs. However, with higher burn-ups and power uprates, it's likely that SCC will become important for both.

Randy Lott, Westinghouse

C: It is important to define the goal before you select the model rather than let the model define the goal. For fuels-related models, development of improved materials and improved fuel designs are probably the high payoff items. For Topical Area 1, the operative mechanisms change drastically from alloy to alloy. Need to have a goal for this study.

A: *[King]* I agree with this comment and am pursuing the development of the goal.

Stuart Maloy, LANL

Q: What was behind the shift from using ion beams as a tool for radiation effects studies to a UQ mission?

A: *[King]* The goals of the initiative are to (1) develop time and length scale transcending models that predict material properties using uncertainty quantification to effectively integrate theory, simulation, and modeling with high dose experimental capabilities, a methodology that has been highly successful in the stockpile stewardship program, and (2) design and develop new radiation tolerant materials using the knowledge gained and methodologies created to shorten the development and qualification time and reduce cost. UQ is not a mission, it is a management tool that effects the integration of theory, simulation, and modeling with ion beam irradiation and it enables assessment of uncertainties when extrapolating to regions where data is limited or nonexistent.

Stuart Maloy, LANL

C: In the clad materials program, we are striving to enable higher burn-up in LWR and fast reactor fuels through developing improved radiation-tolerant cladding materials. In the LWR fuels, this requires qualifying cladding materials out to doses up to 20–50 dpa. In the fast reactor (FR) fuels, this requires qualifying cladding materials out to doses up to 400–450 dpa. Irradiating light water reactor cladding materials in LWRs will take at least 2 to 5 years to perform these irradiations to these doses and 1 to 2 more years afterwards to perform the PIE. Irradiating fast reactor cladding materials in FRs will take 10 to 20 years and 1 to 2 more years afterwards to perform the PIE. If we must wait 5 to 10 years to obtain irradiation data on prospective cladding materials, the materials development is greatly hindered. Ion-irradiation facilities can irradiate prospective cladding materials to such doses in days to months, but the cascades differ from that in a reactor in size and density/rate. These differences have a significant effect on the microstructural development under irradiation and therefore the resulting mechanical properties, but the defects formed at the atomic scale are similar. Thus, it is essential that ion-irradiation results are coupled with appropriately scaled models to understand the differences in flux rate on the resultant microstructure. Such a coupled program with ion-irradiation testing and modeling would not replace long-term reactor irradiations but could help point us to the most radiation-tolerant materials based on fundamental studies of radiation effects in these materials.

Presently, ion-irradiation testing is being used in the clad materials program through use of the LANL ion-beam materials laboratory, ion-irradiation facilities at the University of Wisconsin and the University of Michigan, and soon at LLNL. Modeling studies to complement these ion-irradiation studies are being performed at the University of California at Berkeley in connection with some of the ion-irradiation studies. I believe the modeling studies could be expanded and better coordinated with all of these studies. Perhaps this could be done through direction of NEAMS research or additional work packages under clad materials with additional funding. As the modeling studies are expanded, the ion-irradiation studies will have more meaning and be able to be expanded as well. [Possible candidate materials include] zircaloys for LWRs and ferritic-martensitic steels for FRs.

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Appendix C: Workshop Participants

Name	Affiliation	Name	Affiliation
Todd Allen	University of Wisconsin	Meimei Li	ANL
Tom Arsenlis	LLNL	Randy Lott	Westinghouse
Graham Bench	LLNL	Pete Lyons	Nuclear Energy
Carl Beyer	PNNL	Stuart Maloy	LANL
Mike Billone	ANL	Bill McLean	LLNL
Mark Bourke	LANL	Donn McMahon	LLNL
Luke Brewer	SNL	Gene Nardella	SC
Vasily Bulatov	LLNL	Mike Nastasi	LANL
Mike Burke	Westinghouse	Ken Natesan	ANL
Jeremy Busby	ORNL	Ron Omberg	PNNL
Charlie Davis	Naval Reactors	Peter Pappano	ORNL
Tomas Diaz de la Rubia	LLNL	Kemal Pasamehmetoglu	INL
Mike Dubberly	QuesTek	James Peltz	DOE
Michael Fluss	LLNL	Eric Pitcher	LANL
Frank Garner	TechSource	John Sarrao	LANL
Dave Gelles	PNNL	Bob Schleicher	GA
Bill Goldstein	LLNL	Rebecca Smith-Kevern	Nuclear Energy
John Hack	Bettis Atomic Power Lab	Ron Stambaugh	GA
Bill Halsey	LLNL	Roger Stoller	ORNL
Tony Hill	INL	Robert Tregoning	NRC
Gerard Hofman	ANL	Patrice Turchi	LLNL
Les Jardine	HPDT	Cetin Unal	LANL
Rory Kennedy	INL	Thomas Ward	TechSource
Moe Khaleel	PNNL	Gary Was	University of Michigan
Hussein Khalil	ANL	Brad Williams	DOE
Wayne King	LLNL	Brian Wirth	UC Berkeley
Mark Kirk	ANL	Richard Wright	INL
Richard Klein	LLNL	Abdellatif Yacout	ANL
Rick Kurtz	PNNL	Xinghang Zhang	Texas A&M University
Sue Lesica	Nuclear Energy		

Appendix D: Facilities and Other Resources

Modeling

Name and Location	Pertinent Capability	Availability
Continuum engineering codes: LLNL, SNL, LANL	Large-scale multi-resolution modeling of engineering components with microstructure-aware constitutive functions	Require functionality extensions
Paradis: LLNL DDD: Karlsruhe, Germany	Large-scale Dislocation Dynamics simulations of stress-strain response of irradiated materials	Require functionality extensions
Rate Theory codes: ORNL, LLNL, UCB, UCLA	Mean-field simulations of damage accumulation in irradiated materials	Require extension to complex materials
FPKMC code: LLNL OKMC code: EDF France SPPARKS code SNL	Spatially resolved Monte Carlo simulations of damage accumulation in irradiated materials	Ready to use
MPALE code SNL	Massively parallel, material point method for coupled microstructural evolution and mechanics simulations	
Lattice KMC and mean-field codes: CEA, LLNL	Spatially resolved on-lattice simulations of kinetics in driven multi-component alloys	Requires efficiency enhancements
AMD: LANL Meta-dynamics: ETH	Accelerated atomistic simulations of damage production and annealing in collision cascades. Short-term kinetics in driven alloys.	Can be implemented within short time
Large-scale MD: LLNL, LANL, SNL	Atomistic simulations of displacement damage source databases	Ready to use
Image simulators: PSI, ETH	Simulations of TEM images of irradiation damage microstructure	Available through collaboration
Ab initio codes: ORNL, LLNL, LANL, SNL	Calculations of phase stability, defect energies and diffusivities in multi-component materials	Ready to use

Reactors

Name and Location	Pertinent Capability	Availability
Advanced Test Reactor, Idaho Falls	Irradiation positions for fuels and materials irradiations, short-time irradiations, gamma irradiator.	Available via peer reviewed process (NSUF) or via recharge
MIT Reactor, Boston	Irradiation positions for fuels and materials irradiations, short-time irradiations. Beam tubes, fission converter.	Available via peer reviewed process (NSUF) or via recharge
High Flux Isotope Reactor, Oak Ridge	Irradiation positions for fuels and materials irradiations, short-time irradiations, beam tubes for neutron scattering, gamma irradiator.	Beam tubes available via peer reviewed process (BES User Facility). Irradiation positions available via recharge
MURR, Columbia	Irradiation positions for fuels and materials irradiations.	Available via recharge
Other University Research Reactors, across the U.S.	Beam tubes for neutron science, neutron activation analysis.	Available via recharge
International Reactors (BR2, OSIRIS, HFR, JOYO, BN600, BOR-60, Halden, JHR)	Irradiation positions for fuels and materials irradiations, short-time irradiations, beam tubes for neutron scattering.	Available via recharge
Annular Core Research Reactor (ACRR), Sandia	Pulsed neutron reacto capable of instrumented experiments, temperature mechanical loading	Available via recharge

Ion Beam Capabilities

Location	Pertinent Instruments	Availability/Capabilities
University of Michigan (MIBL)	1.7 MV Tandetron, 400 kV Implanter	Available via recharge
University of Wisconsin (CLIM)	1.7 MV Tandem	Available via recharge. Request ion beam expansion to add a combined irradiation/xrd capability on an open beam line. ~$250K
PNNL (EMSL)	3 MV Tandem	Available via peer reviewed process (BER User Facility).
LLNL (CAMS)	10 MV FN tandem, 1.7 MV Tandem	Available though recharge. H, He and heavy ions, 100 MeV fission-fragment-like ions and ability to irradiate actinide samples. Request straightforward expansion to couple the two accelerators to provide simultaneous high-energy dual beam capability. ~$3M
LANL (MST ion beam laboratory)	3 MV Tandem, 200 kV Implanter	Available via recharge. Low energy simultaneous dual beam capability
ANL	2 MV Tandem, 650 KV Implanter, 100-300 kV TEM	HVEM-Tandem dual beam capability
CEA Saclay (France)	3 MV Pelletron, 2.5 MV Van de Graaff, 2.25 MV tandem	Available through recharge. Simultaneous triple ion beam capability
SNL	6MV Tandem with in situ SEM, 3MV Pelletron, 400kV implanter 100kV nano-implanter	Facilities available from Q1-FY11 onwards

PIE and Sample Preparation

Location	Pertinent Instruments and Capabilities
INL (Modern hot analytical facilities are available at INL)	▪ Dual-Beam Focused Ion Beam (FIB) ▪ Microscale X-Ray Diffractometer (MXRD) ▪ Thermal Ionization Mass Spectrometer (TIMS) ▪ Scanning Thermal Diffusivity Microscope (STDM) ▪ Electron Probe Micro-Analyzer (EPMA) ▪ IASCC test rigs ▪ In-Cell tensile tester ▪ In-Cell EDM ▪ Nanoindenter/AFM-Small sample mechanical testing ▪ Automated micro-hardness tester ▪ FEG-STEM ▪ Atom Probe (few in world available for use on radioactive material, none on fuel) ▪ Raman spectroscopy ▪ SEM hot stage
LLNL	▪ FEI 80-300kV Double Corrected, Monochromated Titan ▪ Philips EM300 with capability to observe actinides, nanoSIMS and FIB ▪ Cryogenic ultra-small x-ray scattering cell ▪ Picoscan AFM systems ▪ Ultraviolet Photoemission Spectroscopy ▪ Confocal time resolved photo-luminescence imaging ▪ Ion Beam Sputter Coating Facility ▪ Mechanical Property Evaluation—mechanical strength, hardness, ductility, toughness, and fracture of materials, components, and assemblies under various conditions of load, strain rate, temperature, and corrosion ▪ Small-Scale, Actinide, Sample Preparation Facility with equipment for research and engineering testing of nuclear materials ▪ Nanoscale Synthesis and Characterization Laboratory with arc melting furnaces, vacuum melting and annealing furnaces, hot–cold rolling, and hot isostatic pressing for powder process ▪ Alloy Synthesis and Thermomechanical Processing
University of Wisconsin, Materials Science Center	▪ Philips CM200 UT TEM, LEO 912 TEM ▪ LEO 1530 FESEM, JEO JSM-6100 SEM, Zeiss 1500XB SEM, Zeiss 1500XB CrossBeam Workstation SEM ▪ Perkin Elmer PHI Model 670 Scanning Auger Microscope ▪ PANalytical X’Pert PRO XRD, Rigaku Small-angle X-ray Scattering System ▪ Stoe X-ray diffractometer, PANalytical X-ray Diffractometer ▪ AFM ▪ Aramis Confocal Raman Microscope, ▪ Perkin Elmer 5400 ESCA Spectrometer

Location	Pertinent Instruments and Capabilities
ORNL Shared Research Equipment (SHaRE) User facility The Irradiated Fuels Examination Laboratory (IFEL) Irradiated Materials Examination and Test Lab (IMET) The Low Activation Materials Development and Analysis (LAMDA) facility	▪ FEI Titan S 80-300 TEM/STEM with CEOS probe-corrector and EELS ▪ Hitachi HF3300 FEG-TEM/STEM with EDS ▪ FEI/Philips CM200 FEG-TEM/STEM with EDS, EELS/EF-TEM ▪ JEOL 6500 FEG-SEM with SDD-EDS and OIM/EBSD, Philips XL30 SEM ▪ Hitachi NB5000 Dual Beam FIB with SDD-EDS and OIM/EBSD ▪ Local Electrode Atom Probe (LEAP) ▪ Imago Scientific Instruments Laser-LEAP ▪ FEI Nova 200 Dual Beam FIB with EDS ▪ Additional EM Specimen Preparation Facilities ▪ Laser Profilometer, Precision densitometer ▪ Instron tensile machine with high vacuum chamber ▪ Automated ball indentation system ▪ Automated microhardness indenter ▪ Instrumented Charpy impact system ▪ Computer-controlled fracture toughness and fatigue systems. ▪ Annealing furnace, ▪ CNC milling machine ▪ Testing with Instron tensile machine
LANL	▪ FEI Titan ▪ FEI Tecnai F30 Analytical TEM/STEM ▪ JEOL 3000F High Resolution TEM ▪ FEI Strata DB235 FIB/SEM ▪ FEI XL30 Environmental SEM and Orientation Imaging System ▪ Helios FIB, In-cell tensile tester, in-cell milling machine, In-cell band saw, In-cell sample preparation Leitz MM5 optical microscope with hardness tester in cell ▪ Plutonium facility, TA-55, PF-4 Cat I Nuclear facility for production of ceramic fuels ▪ Materials Science Laboratory MSL, TA-3, 1698 Gloveboxes for production of uranium based fuels with depleted uranium ▪ Sigma building, TA-3: hot (radioactive) FIB for preparation of TEM foils from irradiated materials, hot (radioactive) EDM for preparation of mechanical test specimens from irradiated materials, forging and rolling capabilities, hot isostatic pressing capabilities
UNLV	▪ FEI tecnai F30 Analytical TEM/STEM for analysis of radioactive materials
University of Michigan (MIBL)	▪ Ion Beam Assisted Deposition System (IBAD) ▪ Hardness Indenter ▪ Vacuum Furnace ▪ Dektak 3 profilometer ▪ Residual Stress Measurement System

Location	**Pertinent Instruments and Capabilities**

SNL	▪ Electron Microscopy
	  ▪ Transmission electron microscopy (TEM)
	    ▪ FEI Tecnai 300keV TEM/STEM with EDX and EELS capabilities
	    ▪ Philips CM30 300keV TEM with EDX and EELS capabilities
	    ▪ New aberration-corrected STEM with EDX and EELS capabilities being purchased, available Q2 FY11
	    ▪ In situ liquid and gas cell capabilities coming on line.
	  ▪ Scanning Electron Microscopy (SEM)
	    ▪ 2 FEG SEM's (Zeiss Supra 55 VP-FEG-SEM) with EDX and electron backscattered diffraction (EBSD),
	    ▪ 1 FEG SEM (FEI Magellan) with ultra-high resolution, (0.8nm)
	  ▪ Dual Electron-Focused Ion Beam (FIB) Instruments
	  ▪ FEI DB235 FIB for TEM sample preparation and micropillar fabrication
	  ▪ 2 FEI Helios Nanolab instruments-dual beam FIB with EBSD
	▪ Mechanical Characterization
	  ▪ 3 nanoindentation instruments
	    ▪ Hysitron TI-900 for fully automated indentation measurement with displacement control
	    ▪ Nanoinstruments XP for higher-load instrument indentation
	    ▪ Micromaterials Nanotest and Microtest Instruments allow for elevated temperature indentation
	  ▪ Full mechanical testing suite including:
	    ▪ Traditional mechanical testing load frames up to temperatures of 2000°C and up to strain rates of 100/s
	    ▪ Special testing equipment for MEMS and other micro and miniature scale geometries
	    ▪ Equipment and expertise for non-contact, digital image correlation-based mechanical characterization

User Facilities

Location	Pertinent Instruments and Capabilities
Stanford Synchrotron Radiation Laboratory, Stanford	Beamline 8.2 at the Stanford Synchrotron Radiation Laboratory (SSRL) uses bend-magnet radiation and a spherical grating monochromator. X-ray near-edge absorption spectroscopy (XANES) is routinely measured through three methods that can differentiate between surface and bulk properties of nanoscale materials. Total electron yield (TEY) detection can probe to about 5 nanometers, while Auger electron yield (AEY) detection is more surface-sensitive, probing the outermost area (~ 1 nm). Bulk sensitivity is gained through total fluorescence yield (TFY). Experiments can also be used for high-resolution x-ray photoelectron spectroscopy (XPS) to further characterize materials.
Advanced Photon Source, ANL	The ultra-small-angle x-ray scattering (USAXS) end station at sector 32 of the Advanced Photon Source acquires scattering from 10^{-4} Å^{-1} to 1 Å^{-1}, covering four orders of magnitude in scattering angle and capable of covering approximately nine orders of magnitude in intensity. This technique can be used to study structures from a few nanometers to a few micrometers in size and is perfectly suited to study nanoporous structures. Size distributions for the minority phase can be extracted, along with the shape of the scattering population, whether spherical (three-dimensional), disk-like (two-dimensional), or rod-like (one-dimensional).
National Center for Electron Microscopy (NCEM), LBNL	NCEM features several unique instruments, complemented by strong expertise in computer image simulation and analysis. NCEM also maintains one-of-a-kind instruments for imaging magnetic materials and develops techniques and instrumentation for dynamic in situ experimentation. http://ncem.lbl.gov/frames/center.htm
Los Alamos Neutron Science Center, LANL	http://lansce.lanl.gov

LLNL Computational Resources

Name and Location	Pertinent Capability	Availability
Livermore Computing (LC), LLNL	LLNL's premier high-performance computing organization serving scientists and engineers. LC is a full service computing organization serving a large customer base (over 2,600 active users; 840 are off-site) with computing, storage, data management, and visualization services.	LC services are offered through the Multiprogrammatic and Institutional Computing Program.
Green Data Oasis, LLNL	A large data store (620 TB) intended to facilitate the sharing of scientific data with external collaborators. Solaris containers (virtualization): gives each project their own IP address, services, quotas.	The Green Data Oasis is managed through the Multiprogrammatic and Institutional Computing Program.
Access Grid Nodes, LLNL	The Access Grid® is a multi-media interactive environment used to support group-to-group interactions across the Grid.	LLNL has one node in B-451 that is being upgraded and is installing a second node in B-435. Both will be available to M&IC customers.
PowerWalls, LLNL	High-resolution displays for work group collaborations, presentations to large audiences, and program reviews.	LLNL has three PowerWalls for use by M&IC customers.

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